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RADIONUCLIDE ANALYSIS BY GAMMA SPECTROSCOPY

Public Health Service
Washington, D. C.

1973

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presented by:

Training Branch
Division of Radiological Health

NATIONAL TECHNICAL
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U. S. DEPARTMENT OF HEALTH,
EDUCATION, AND WELFARE

Public Health Service

342

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Training Branch
Division of Radiological Health

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Bureau of State Services--Environmental Health
Washington, D. C. 20201

Foreword

The Division of Radiological Health has been designated by the Public Health Service as the focus of a national program to plan, conduct, and coordinate efforts toward the prevention and control of radiation hazards to public health so that the Nation may obtain maximum benefits of radiation with minimum risk to health. The Division assists State and local health agencies and others concerned with radiation protection, conducts research and field investigations, maintains surveillance to determine levels of radiation in the environment, and conducts or coordinates projects of a special nature carried out in conjunction with other Federal agencies.

For more than a decade, the Public Health Service has been training professional personnel engaged in health and health-related work in the principles and techniques needed in long-range programs for radiation protection and control. The cornerstone of this training effort has been technical short courses of from 1- to 3-weeks' duration presented by radiation health specialist instructors of the Division. Currently, the principal training centers are located at the Radiological Health Laboratory, Rockville, Maryland, the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, and the Southwestern Radiological Health Training Laboratory, Las Vegas, Nevada. Courses are also scheduled in the field at the Northeastern Radiological Health Laboratory, Winchester, Massachusetts, the Southeastern Radiological Health Laboratory, Montgomery, Alabama, and by special arrangements in individual States.

Our training goal is to provide health personnel, not only with an understanding of the effect on individuals and the general population of exposure to any source of ionizing radiation, but also with broad principles and practical methods of control. We have achieved our objective in part if this manual, prepared by our training instructors, in anyway contributes to professional or public knowledge of how to prevent or minimize radiation hazards.

Donald R Chadwick

Donald R. Chadwick, M.D.
Chief, Division of Radiological Health

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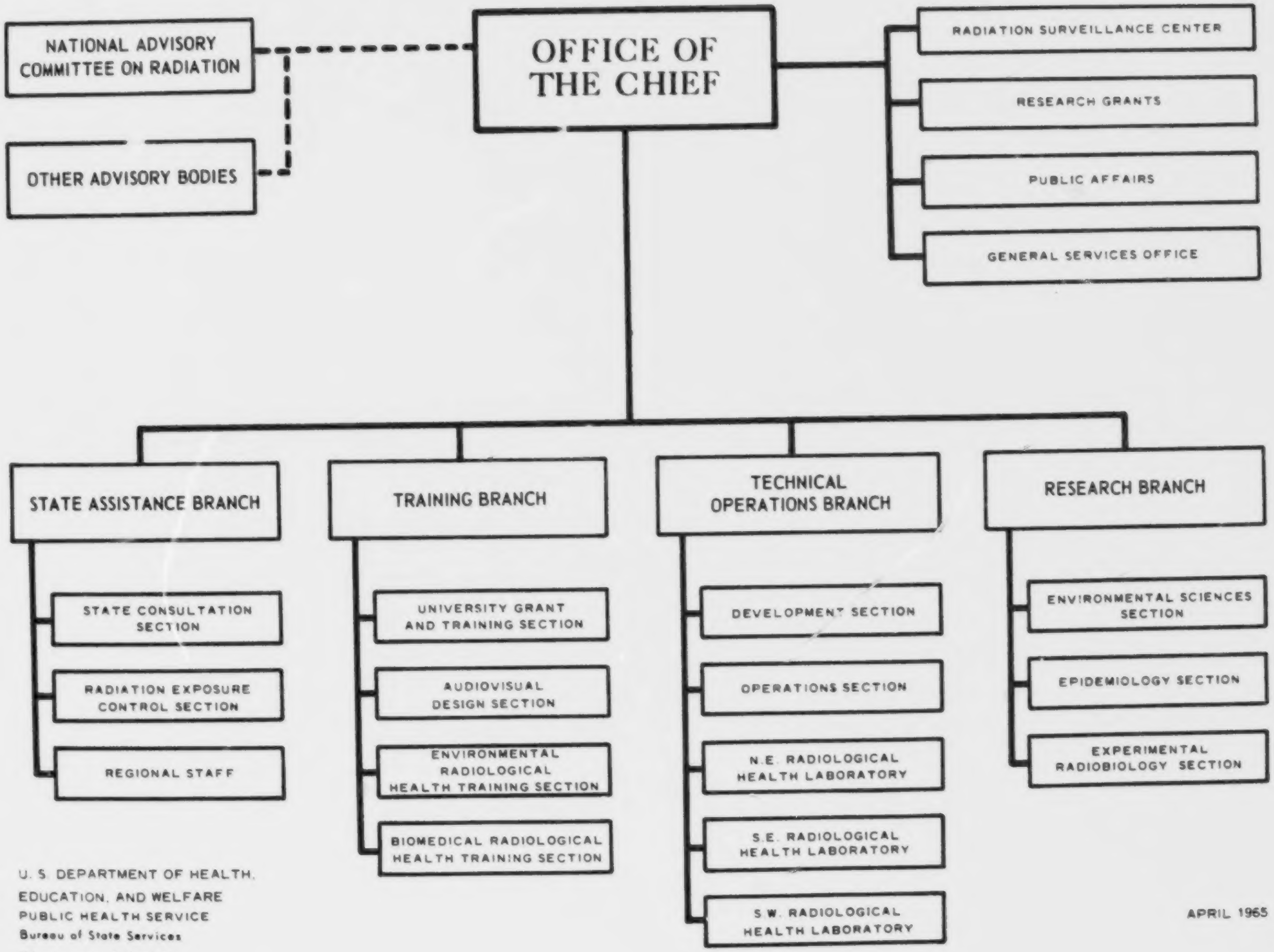
DIVISION OF RADIOLOGICAL HEALTH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Sources of Information in Radiological Health

I. INTRODUCTION

There are many sources of information available to personnel working in the field of radiological health. The purpose of this report is to list the variety of materials available and to outline procedures for obtaining them.

II. TEXTBOOKS

Textbooks provide some valuable basic information in the field of radiological health, though some may deal exclusively with atomic and nuclear physics, others with nomenclature and units, still others with radiation-generating machines and radioactive materials --- their use, and the design of facilities for managing them.

Listed here are some good basic reference texts:

A. Units, Nomenclature, and Basic Data

1. American Standard ASA N1. 1-1957, "Glossary of Terms in Nuclear Science and Technology," American Society of Mechanical Engineers, 29 West 39th Street, New York 18, N.Y. 1957, $5.

2. Glasstone, Samuel, "Source Book on Atomic Energy," D. Van Nostrand Co., Inc. 257 Fourth Avenue, New York 10, New York, 1958. $4.40.

3. Kinsman, Simon, editor, "Radiological Health Handbook", (Revised) Publication No. PB-121784R, U.S. Department of Commerce, Office of Technical Services, Washington 25, D.C. 1960. $3.75.

4. Blatz, Hanson, editor-in-chief, "Radiation Hygiene Handbook," McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y. 1959. $27.50.

5. Etherington, Harold, editor, "Nuclear Engineering Handbook," first edition, McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y. 1958. $25.00.

6. Dummer, Jerome E., Jr., "Handbook for Radiation Monitoring," LA-1835, Third Edition, Los Alamos Scientific Laboratory of the University of California, Los Alamos, New Mexico. For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., 65 cents.

B. Atomic and Nuclear Physics

1. Lapp, R. E., and Andrews, H. L., "Nuclear Radiation Physics," second edition, Prentice-Hall, Inc., 70 Fifth Ave, New York 11, N.Y. 1954. $10.

2. Semat, Henry, "Introduction to Atomic and Nuclear Physics," Rinehart & Co., 232 Madison Ave, New York 16, N.Y., 1954. $6.50.

3. Kaplan, Irving, "Nuclear Physics," Addison-Wesley Publishing Co., Inc., 500 Kendall Square Building, Cambridge 42, Mass., 1955. $9.75.

C. Instrumentation and Assay Techniques

1. Friedlander, Gerhart, and Kennedy, J. W., "Nuclear and Radiochemistry," John Wiley & Sons, Inc., 440 Fourth Avenue, New York 16, N.Y., 1955. $7.50.

2. Comar, C. L., "Radioisotopes in Biology and Agriculture -- Principles and Practice," McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y., 1955. $9.

3. Overman, R. T., and Clark, H. M., "Radioisotope Techniques," McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y., 1959.

4. Faires, R. A., and Parks, B. H., "Radioisotope Laboratory Techniques," Pitman Publishing Corp., 2 West 45th Street, New York 36, N.Y., 1958. $5.75.

5. Korff, S. A., "Electron and Nuclear Counters: Theory and Use," D. Van Nostrand Company, Inc., 257 Fourth Avenue, New York 10, N. Y., $6.75.

6. Oak Ridge National Laboratory, "Master Analytical Manual," TID-7015 Technical Information Extension, U.S. Atomic Energy Commission, Oak Ridge, Tenn., 1958. $9.

D. Nuclear Engineering

1. Smyth, H. D., "A General Account of the Development of Methods of Using Atomic Energy for Military Purposes Under the Auspices of the United States Government (1940-1945)," Available from Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., 1945. 40 cents.

2. Murray, R. L., "Introduction to Nuclear Engineering," Prentice-Hall, Inc., 70 Fifth Avenue, New York 11, N.Y., 1954, $11.35.

3. Stephenson, Richard, "Introduction to Nuclear Engineering," McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y., 1954. $9.50.

4. Glasstone, Samuel, "Principles of Nuclear Reactor Engineering," D. Van Nostrand Co., Inc., 257 Fourth Avenue, New York 10, N.Y., 1955. $7.95.

5. McCullough, C. R., "Safety Aspects of Nuclear Reactors," D. Van Nostrand Co., Inc., 257 Fourth Avenue, New York 10, N.Y., 1957. $8.50.

E. X-ray Technology

1. Files, Glenn W., editor, "Medical Radiographic Technic", second edition, Charles C. Thomas, Publisher, Springfield, Ill., 1959. $11.00.

2. Sarsfield, Leslie, G. H., "Electrical Engineering in Radiology", Instruments Publishing Co., Pittsburg, Penn., 1936.

3. Sproull, Wayne, T., "X-ray in Practice", McGraw Hill, New York, 1946, $12.50.

4. Robertson, J. K., "Radiology Physics", D. Van Nostrand Co., Inc., New York, 1948.

5. Selman, Joseph, "The Fundamentals of X-ray and Radium Physics", second edition, Charles C. Thomas, Publisher, Springfield, Ill., 1958. $8.50.

F. Radiation Protection

1. Braestrup, C. B., and Wyckoff, H. O., "Radiation Protection," Charles C. Thomas, Publisher, 301 East Lawrence Avenue, Springfield, Ill., 1958. $10.50.

2. Sonnenblick, B. P., editor, "Protection in Diagnostic Radiology," Rutgers University Press, 30 College Avenue, New Brunswick, N.J., 1959. $7.50.

3. Goldstein, Herbert, "Fundamental Aspects of Reactor Shielding," Addison-Wesley Publishing Co., Inc., Reading, Mass., 1959. $9.75.

4. "Radiation Protection Criteria and Standards: Their Basis and Use", Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy,

Congress of the U.S., 86th Congress, 2nd Session 1960. Available from U.S. Government Printing Office.

G. Radiobiology and Medicine

1. Quimby, E. H., Feitelberg, S., and Silver, S., "Radioactive Isotopes in Clinical Practice," Lea and Febiger, Publishers, 600 Washington Square, Philadelphia 6, Pa., 1958. $10.

2. Hollaender, Alexander, editor, "Radiation Biology", McGraw-Hill Book Co., Inc., 330 West 42nd Street, New York 36, N.Y., Vol. I 1954. two parts, $18; Vol. II 1955. $8.50; Vol. III 1956, $10.50.

3. Blahd, William H., Baver, Franz K., and Classon, Benedict, "The Practice of Nuclear Medicine", Charles C. Thomas, Publisher, Springfield, Ill., 1958. $12.50.

4. Behrens, C. F., "Atomic Medicine," third edition, Williams & Wilkins Co., 428 East Preston Street, Baltimore 2, Md. 1959. $15.00.

5. Andrews, G. A., Brucer, M., and Anderson, E. B., editors, "Radioisotopes in Medicine," Report ORO 125, September, 1953. For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., $5.50 paper cover.

6. Bacq, Z. M., and Alexander, Peter, "Fundamentals of Radiobiology", Academic Press, 111 Fifth Avenue, New York 3, N.Y. 1955. $6.50.

7. Lacassagne, Antonie, and Gricouroff, Georges, "Action of Radiation on Tissues", second edition, Grune & Stratton, Inc., 381 Fourth Avenue, New York 16, N.Y., 1958. $6.25.

H. Nuclear Weapons

1. Glasstone, Samuel, editor, "The Effects of Nuclear Weapons", prepared by the U.S. Department of Defense and published by the U.S. Atomic Energy Commission, June 1957. For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., $2.00.

2. The Nature of Radioactive Fallout and Its Effects on Man", Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the U.S., 85th Congress, 1st session 1957. Available from U.S. Government Printing Office.

I. Radioactive Wastes

1. "Industrial Radioactive Waste Disposal", Hearings before the special Radiation Subcommittee of the Joint Committee on Atomic Energy, Congress of the United States, 1959. Available from U.S. Government Printing Office.

2. "Radioactive Waste Disposal from Nuclear-Powered Ships", a report of the Committee on the Effects of Atomic Radiation on Oceanography and Fisheries, National Academy of Sciences' Study of the Biological Effects of Atomic Radiation, Publication 658, National Academy of Sciences --- National Research Council, 2101 Constitution Avenue, NW, Washington 25, D.C.

J. Federal-State Relationships

1. "Selected Materials on Federal-State Co-operation in the Atomic Energy Field", report issued by the Joint Committee on Atomic Energy, Congress of the United States, March 1959.

K. General

1. "Proceedings of the International Conference on the Peaceful Uses of Atomic Energy", 16 Volumes, United Nations, New York, 1956.

2. "Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy", 34 Volumes, United Nations, New York 1959. An edited series of 12 volumes is available from Pergamon Press, 122 East 55th Street, New York 22, N.Y. 1959.

3. "Annual Review of Nuclear Science", 8 Volumes, 1952-1958. Annual Reviews, Inc., Palo Alto, California.

4. Caldecott, Richard S., and Snyder, Leon A., editors, "Radioisotopes in the Biosphere", University of Minnesota, Center for Continuation Study, 1960.

5. Meneely, George R., "Radioactivity in Man", Charles C. Thomas, Publisher, Springfield, Ill., 1961. $16.50.

III. CURRENT LITERATURE

Public health workers having responsibilities in the area of radiological health must keep abreast of current literature in a rapidly changing field. The references below represent titles and sources for some of the more recent publications of this type, with the suggestion that the same sources may be providing still newer information from time to time.

A. Recommendations of the National Committee on Radiation Protection and Measurements (NCRP) published as Handbooks of the National Bureau of Standards. Available from the Superintendent of Documents, Government Printing Office, Washington 25, D.C. Titles and prices of the Handbooks include:

1. Handbook 42, "Safe Handling of Radioactive Isotopes", September 1949. 20 cents.

2. Handbook 48, "Control and Removal of Radioactive Contamination in Laboratories", Dec. 15, 1951. 15 cents.

3. Handbook 49, "Recommendations for Waste Disposal of Phosphorous 32 and Iodine 131 for Medical Users", November 2, 1951. 15 cents.

4. Handbook 50, "X-Ray Protection Design", May 9, 1952, 20 cents.

5. Handbook 51, "Radiological Monitoring Methods and Instruments", April 7, 1952. 20 cents.

6. Handbook 53, "Recommendations for the Disposal of Carbon-14 Wastes", October 26, 1953. 15 cents.

7. Handbook 54, "Protection Against Radiations from Radium, Cobalt-60, and Cesium-137", Sept. 1, 1954. 25 cents.

8. Handbook 55, "Protection Against Betatron-Synchrotron Radiations up to 100 Million Electron Volts", Feb. 26, 1954. 25 cents.

9. Handbook 57, "Photographic Dosimetry of X-Rays and Gamma Rays", August 20, 1954. 15 cents.

10. Handbook 58, "Radioactive-Waste Disposal in the Ocean", August 25, 1954. 20 cents.

11. Handbook 59, "Permissible Dose from External Sources of Ionizing Radiation", Sept. 24, 1954. 30 cents.

12. Handbook 61, "Regulation of Radiation Exposure by Legislative Means", Dec. 9, 1955. 25 cents.

13. Handbook 63, "Protection Against Neutron Radiation up to 30 Million Electron Volts", Nov. 22, 1957. 40 cents.

14. Handbook 64, "Design of Free-Air Ionization Chambers", Dec. 13, 1957. 20 cents.

15. Handbook 65, "Safe Handling of Bodies Containing Radioactive Isotopes", July 10, 1958. 15 cents.

16. Handbook 66, "Safe Design and Use of Industrial Beta-Ray Sources", May 28, 1958. 20 cents.

17. Handbook 69, "Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure", June 5, 1959. 35 cents.

18. Handbook 72, "Measurement of Neutron Flux and Spectra for Physical and Biological Applications", July 15, 1960. 35 cents.

19. Handbook 73, "Protection Against Radiations from Sealed Gamma Sources", July 27, 1960. 30 cents.

20. Handbook 76, "Medical X-ray Protection up to 3 Million Volts", 1961. 25 cents.

21. Handbook 78, "Report of the International Commission on Radiological Units and Measurements", 1959. 65 cents.

B. Publications of the Federal Radiation Council, a statutory body charged with the responsibility for advising the President with respect to radiation matters directly or indirectly affecting health, including the guidance of all Federal agencies in the formulation of radiation standards and in the establishment and execution of programs of cooperation with States:

1. Background Material for the Development of Radiation Protection Standards, Report No. 1, May 13, 1960. For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., Price 30 cents.

C. Publications of the United States Atomic Energy Commission

1. Nuclear Science Abstracts, a publication of the AEC Technical Information Service Extension. Available from the Superintendent of Documents, Government Printing Office, Washington 25, D.C., $7.50 per year.

This report, issued twice monthly, contains abstracts of articles of interest in all areas of nuclear science and technology. In particular, it includes summaries of all nonclassified reports issued by the various AEC installations. Copies of the complete reports are available on loan from AEC Depository Libraries which are located as follows:

AEC DEPOSITORY LIBRARIES

ALABAMA
  Auburn, Alabama Polytechnic Institute Library
  Birmingham, Birmingham Public Library

ARIZONA
  Tucson, University of Arizona

ARKANSAS
  Fayetteville, University of Arkansas Library

CALIFORNIA
  Berkeley, University of California General Library
  Los Angeles, University of California Library
  Menlo Park, Stanford Research Institute
  San Diego, San Diego Public Library

COLORADO
  Denver, Denver Public Library
  The City and County of Denver

CONNECTICUT
  New Haven, Yale University Library

DELAWARE
  Newark, University of Delaware

DISTRICT OF COLUMBIA
  Washington, Library of Congress

FLORIDA
  Coral Gables, University of Miami Library
  Gainesville, University of Florida Library

GEORGIA
- Atlanta, Georgia Institute of Technology Library

HAWAII
- Honolulu, University of Hawaii

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- Chicago, John Crerar Library
- Chicago, University of Chicago Library
- Evanston, Northwestern University Library
- Urbana, University of Illinois Library

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- Indianapolis, Indianapolis Public Library
- Lafayette, Purdue University Library

IOWA
- Ames, Iowa State College Library

KANSAS
- Manhattan, Kansas State College School of Engineering and Architecture

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- Lexington, University of Kentucky Library
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- St. Louis, Washington University

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- Albuquerque, University of New Mexico Library

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- Buffalo, Lockwood Memorial Library, University of Buffalo
- Ithaca, Cornell University Library
- New York, Atomic Industrial Forum
- New York, Columbia University
- New York, New York Public Library
- Rochester, University of Rochester
- Schenectady, Union College Library
- Syracuse, Syracuse University
- Troy, Renselaer Polytechnic Institute Library

NORTH CAROLINA
- Charlotte, Charlotte and Mechklenburg County Public Library
- Durham, Duke University Library
- Raleigh, North Carolina State College Library

OHIO
- Cincinnati, University of Cincinnati
- Cleveland, Cleveland Public Library
- Columbus, Ohio State University
- Toledo, University of Toledo Library
- Youngstown, Youngstown University

OKLAHOMA
Stillwater, Oklahoma State University Library

OREGON
Corvallis, Oregon State College
Portland, Portland Public Library

PENNSYLVANIA
Philadelphia, University of Pennsylvania Library
Pittsburgh, Carnegie Library of Pittsburg
University Park, Pennsylvania State University Pattee Library

PUERTO RICO
Rio Piedras, University of Puerto Rico Main Library

RHODE ISLAND
Providence, Brown University

SOUTH CAROLINA
Columbia, University of South Carolina, McKissick Memorial Library

TENNESSEE
Knoxville, University of Tennessee
Memphis, Cossett Memorial Library
Nashville, Joint University Libraries

TEXAS
Austin, University of Texas
College Station, Agricultural and Mechanical College of Texas
Dallas, Southern Methodist University
Houston, Rice Institute
San Antonio, San Antonio Public Library

UTAH
Salt Lake City, University of Utah Library

VIRGINIA
Blacksburg, Virginia Polytechnic Institute Library
Charlottesville, University of Virginia, Alderman Library

WASHINGTON
Pullman, State College of Washington
Seattle, University of Washington

WEST VIRGINIA
Morgantown, West Virginia University Library

WISCONSIN
Madison, University of Wisconsin Library
Milwaukee, Milwaukee Public Library

WYOMING
Laramie, University of Wyoming.

2. Reports of the U.S. Atomic Energy Commission to Congress (semiannual; annual beginning with report for all of 1959), available from the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C. Each of the later reports has an alternate title indicating the principal subject. Examples are:

a. First Semiannual Report, January 1947.

b. Sixth Semiannual Report, "Atomic Energy and the Life Sciences", July 1949. 45 cents.

c. Thirteenth Semiannual Report, "Assuring Public Safety in Continental Weapons Tests", January 1953. 50 cents.

d. Twenty-first Semiannual Report, "Radiation Safety and Major Activities in the Atomic Energy Programs", January 1957. $1.25.

e. Twenty-fifth Semiannual Report," "Atomic Industrial Progress and Second World Conference", January 1959. $1.25.

f. Twenty-sixth Report, "Major Activities in the Atomic Energy Programs", emphasizing management of radioactive wastes, January 1960. $2.00.

g. "Major Activities in the Atomic Energy Programs - January - December 1960", Annual Report to Congress of the U.S. Atomic Energy Commission for 1960, January 1961. $1.75.

3. Special Reports

The Atomic Energy Commission issues a number of special reports from time to time which are very useful in radiological health work. They may be obtained without charge from the Technical Information Service Extension, P.O. Box 1001. Oak Ridge, Tennessee. Examples are:

a. "Guide to Atomic Energy Literature for the Civilian Application Program", TID-4575 (First revision) Nov. 1957.

b. "What's Available in the Unclassified Atomic Energy Literature", TID-4550 (Fifth revision) July 1958.

c. "Special Sources of Information on Isotopes", TID-4563 September, 1957.

d. "Technical Books", 1947 to 1959.

C. Reports of the National Academy of Sciences - National Research Council, 2101 Constitution Avenue NW, Washington 25, D.C. For example:

1. "The Biological Effects of Atomic Radiation - A Report to the Public", 1960.

2. "The Disposal of Radioactive Waste on Land", report of the Committee on Waste Disposal of the Division of Earth Sciences, April 1957.

3. "Pathological Effects of Atomic Radiation", National Research Council Publication #452. 1956. $1.00.

4. "Measurements and Standards of Radioactivity", National Research Council Publication #573. 1958.

D. Reports of the American Public Health Association, Inc., 1790 Broadway, New York 19, N.Y.

For example:

1. "Public Exposure to Ionizing Radiations - What Public Health Personnel Need to Know", 1958.

E. Reports of The American College of Radiology, 20 North Wacker Drive, Chicago 6, Illinois.

For example:

1. "A Practical Manual on the Medical and Dental Use of X-rays With Control of Radiation Hazards", 1958.

F. Reports of the National Fire Protection Association, 60 Batterymarch Street, Boston 10, Mass.

For example:

1. "Recommended Safe Practice for Laboratories Handling Radioactive Materials", May 1955. 50 cents.

G. Reports of the United Nations, New York.

For example:

1. "Report of the United Nations Scientific Committee on the Effects of Atomic Radiations", Thirteenth Session, Supplement No. 17, (A/3838) 1958. $2.50.

H. Reports of the International Atomic Energy Agency, Karnter Ring, Wien I, Austria.

For example:

1. "Safe Handling of Radioisotopes", Safety Series No. I, 1958. $1.00.

I. Reports of the American Hospital Association, 18 East Division Street, Chicago 10, Illinois.

For example:

1. "Manual on the Use of Radioisotopes in Hospitals".

J. Periodicals

1. Radiological Health Data, issued monthly by the Public Health Service in conjunction with the responsibility of this agency

for the collation, analysis, and interpretation of data on environmental radiation levels. For sale by the U.S. Department of Commerce, Office of Technical Services, Washington 25, D.C. Price $9.00 per year.

2. Health Physics, the Official Journal of the Health Physics Society, Pergamon Press, Inc., 22 East 55th Street, New York 22, N.Y. $17.00 per year.

3. Nucleonics, published monthly by the McGraw-Hill Publishing Co., Inc., 330 West 42nd Street, New York 36, New York, $8.00 per year.

4. Science, published weekly by American Association for Advancement of Science, 1515 Massachusetts Avenue, N.W., Washington, D.C., $8.50 per year.

5. Public Health Engineering Abstracts, published monthly by the U.S. Department of Health, Education, and Welfare, Public Health Service. Available from Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C., $2.00 per year.

6. Bulletin of the Atomic Scientists, a magazine for science and public affairs, published monthly by the University of Chicago Press, 5750 Ellis Avenue, Chicago 37, Ill., $6.00 per year.

7. Radiology, published monthly by the Radiological Society of North America, 713 East Genessee Street, Syracuse 2, N. Y., $10.00 per year.

8. American Journal of Roentgenology, Radium Therapy, and Nuclear Medicine, the official organ of The American Roentgen Ray Society and The American Radiation Society, published monthly by Charles C. Thomas, 301 - 327 East Lawrence Ave., Springfield, Ill., $15.00 per year.

9. The Forum Memo, a monthly report to members of the Atomic Industrial Forum, Inc., 3 East 54th Street, New York 22, N.Y. The Forum also publishes special supplements to the Memo dealing with particular developments in nuclear science, proceedings of conferences organized by the Forum, and separate publications covering specific aspects of the field. The latter are on sale to the public.

10. Atomic Industry Reporter, published weekly by the Bureau of National Affairs, Inc., Washington, D.C. A review of all major developments in the field of nuclear science and technology.

IV. TRAINING

Many of the colleges and universities in the United States are incorporating several of the phases of radiological health into existing courses, while others are setting up new courses to cover the subject. As a result, graduates of tomorrow should be well-grounded in radiological health. In the meantime, the majority of public health personnel must acquire necessary fundamentals of radiological health through means of specialized short courses.

A. A number of colleges and universities have assisted in this work through presentation of radiological health seminars, symposia, and short courses. A brief listing of a few recent examples of these is given below, with the suggestion that queries on new programs be directed to the institutions.

1. Two-day conference on "Radioactivity and the Environment", Rutgers University, New Brunswick, N. J., April 9-10, 1959.

2. A one-week course on "Radiation for Industrial Physicians and Lawyers", University of Cincinnati, Cincinnati 10, Ohio, Sept. 9-13, 1957.

3. A three-day course on "Hazards of Atomic Energy", North Carolina State College, Raleigh, N.C., January 1959.

4. A three-day "Seminar on Radiological Health" by Department of Sanitary Engineering, University of North Carolina, Chapel Hill, N.C., January 26-28, 1959.

5. Summer courses on Nuclear Technology at the University of California, Berkeley:

a. "Nuclear Technology Study Program", June 22 - Aug. 28, 1959.

b. "Radiological Regulation and Use", July 20-31, 1959.

c. "Nuclear Technology Survey", July 13-17, 1959.

6. Summer Courses on Radiological Health by the Postgraduate Medical School, New York University, New York, N. Y.

a. "Introduction to Radiological Health", Jan. 5-16, 1959.

b. "Radiological Health Laboratory", Jan. 19-30, 1959.

c. "Radiochemical Analysis", Nov. 10-21, 1959.

d. "Radiation Hygiene Measurements", May 4-29, 1959.

B. Short-term technical training courses are also being offered by industrial groups. Examples are:

1. "Industrial Radiology", St. John X-Ray Laboratory, Califon, New Jersey. This course includes sessions on radiation hazards and safety precautions.

2. Three to five day "Radioisotope Training Courses", Tracerlab, Inc., Waltham, Mass.

C. Short-term technical courses available through the Federal government include those presented by:

1. Special Training Division, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tenn. Typical courses offered by the Institute include:

a. "Radioisotope Research Techniques", two weeks. Laboratory work and appropriate lectures on techniques which will enable participants to undertake research with radionuclide tracers.

b. "Radioisotopes in Industry", a six-week course designed to assist technical personnel in obtaining sufficient facility in the use of radionuclides to utilize them safely and efficiently in their industrial organizations.

2. Division of Radiological Health, Public Health Service, Robert A. Taft Sanitary Engineering Center, 4676 Columbia Parkway, Cincinnnati 26, Ohio.

A total of approximately one dozen different courses are offered under a full-time training program.

These include:

a. "Basic Radiological Health", a two-week course covering the basic technical knowledge essential to radiological health work. The course is directed principally to engineers and scientists.

b. "Medical Aspects of Radiological Health", a two-week basic course for medical, dental, and biological personnel.

c. "Medical X-Ray Protection", a one-week course covering procedures for inspection and survey of x-ray facilities.

d. "Radioactive Pollutants in Water", a one-week course providing technical training for water supply and water pollution control personnel concerned with radioactive contamination of the water environment.

e. "Radionuclides in Foods", a two-week course designed for the professional personnel having responsibilities for surveillance of radioactive materials in milk and food.

V. FILMS

Many films covering one or more of the several phases of radiological health have been produced. These may serve as an excellent source of information and are especially valuable as teaching aids.

A. The Atomic Energy Commission maintains 10 motion picture film libraries in the United States, each stocking almost 150 unclassified 16-mm. films with sound track. A catalog describing these films is available upon request. To request the catalog or borrow a film, write the library serving your area as given below:

If you are from Canada, Maine, New Hampshire, Vermont, Massachusetts, Connecticut, Rhode Island, Pennsylvania, New Jersey, or New York, contact:

Director, Public Information Service
U.S. Atomic Energy Commission
New York Operations Office
376 Hudson Street
New York 14, N.Y.

If you are from Delaware, Maryland, Virginia, West Virginia, or the District of Columbia contact:

Public Information Service (Pictorial)
U.S. Atomic Energy Commission
Washington 25, D.C.

If you are from Alabama, Florida, North Carolina, South Carolina, or Georgia, contact:

Assistant to the Manager for Public Education
U.S. Atomic Energy Commission
Savannah River Operations Office
P.O. Box A, Aiken, S.C.

If you are from Montana, Utah, or Idaho, contact:

Assistant to the Manager for Information
U.S. Atomic Energy Commission
Idaho Operations Office
P.O. Box 1221, Idaho Falls, Idaho

If you are from California or Hawaii, contact:

Assistant to the Manager
U.S. Atomic Energy Commission
San Francisco Operations Office
518 17th Street, Oakland 12, Calif.

If you are from Colorado, Wyoming, Kansas, or Nebraska, contact:

Director, Information Division
U.S. Atomic Energy Commission
Grand Junction Operations Office
Grand Junction, Colo.

If you are from North Dakota, South Dakota, Missouri, Iowa, Ohio, Indiana, Michigan, Minnesota, Wisconsin, or Illinois, contact:

U.S. Atomic Energy Commission
Chicago Operations Office
P.O. Box 59
Lemont, Illinois
Attn: Mr. Howard C. Baldwin, Information Officer

If you are from Kentucky, Mississippi, Arkansas, Louisiana, or Tennessee, contact:

Public Information Officer
U.S. Atomic Energy Commission
Oak Ridge Operations Office
P.O. Box E, Oak Ridge, Tenn.

If you are from Nevada, Arizona, New Mexico, Texas, or Oklahoma, contact:

Director of Information
U.S. Atomic Energy Commission
Albuquerque Operations Office
P.O. Box 5400, Albuquerque, N.M.

If you are from Washington, (State), Oregon, or Alaska, contact:

Director, Information Division
U.S. Atomic Energy Commission
Hanford Operations Office
P.O. Box 550, Richland, Wash.

B. On the following pages is a list of the titles and descriptions of films currently available on loan from ~~Radiological Health Training Activities, Division of Radiological Health, Robert A. Taft Sanitary Engineering Center, Cincinnati 26, Ohio,~~ the source cited.

RADIATION FUNDAMENTALS: POPULAR, TECHNICAL.

1. "A" Is For Atom - 15 minutes; 16 mm; color.

An amusing character called Dr. Atom takes the audience through "Element Town" to explain the atomic structure of the 92 naturally occurring elements. An understandable explanation of the principles of nuclear fission is given in an interesting and entertaining manner. In cartoon animation, the film goes on to explain principles of atomic energy, and how this energy can be applied to peacetime uses in industry, medicine, agriculture, and science. Released by the General Electric Company.

2. Taking the X out of X-Ray - 10 minutes, 16 mm; black and white.

With narration by Coolidge, this film describes the nature and development of x-rays from their discovery by Roentgen to the present day. Included is a description of the mechanisms by which x-rays are produced from both hot and cold cathode tubes. The theory of fluoroscopy and the production of radiographs is covered. Released by the General Electric Company.

3. Fundamentals of Radioactivity (The Radioisotope - Film I - Part 1) - 45 minutes; 16 mm; black and white.

This film opens with a sequence tracing uranium, the raw material of atomic energy, from the prospector to the Atomic Energy Commission. The film shows how uranium changes into other elements through the natural processes of radioactive decay and nuclear fission. Einstein's equation $E = mc^2$ is cited. Mention is made of the atomic bomb and the use of nuclear power for industry. Stable and radioactive isotopes are also discussed. Released by the Armed Forces Institute of Pathology.

4. Fundamentals of Radioactivity (The Radioisotope - Film I - Part 2) - 35 minutes; 16 mm; black and white.

Charts and energy-level diagrams are employed to illustrate the decay of radionuclides. The various radiations resulting from nuclear changes are described in detail. The nuclear reactor is described in terms of fission and moderation. Several scenes show target materials introduced into a typical nuclear reactor and withdrawn as radionuclides. The processing of fission products is also shown. Released by the Armed Forces Institute of Pathology.

5. Properties of Radiation (The Radioisotope - Film II - Part 1) 30 minutes; 16 mm; black and white.

The penetrations of alpha, beta and gamma radiation using Po^{210}, P^{32}, and Co^{60}, respectively, is demonstrated. Characteristic absorption curves are derived. The cloud chamber, electrostatic generator, and beta ray spectrometer are introduced. Concepts of the electron volt, beta ray spectrum, and neutrino are discussed. Released by the Armed Forces Institute of Pathology.

6. Properties of Radiation (The Radioisotope - Film II - Part 2) 38 minutes; 16 mm; black and white.

The continuation of the preceding reel opens with a discussion of ionization, scattering nonlinear absorption, and density thickness (mg/cm^2). In the section concerned with the properties of gamma radiation, the following terms and concepts are introduced: Bremsstrahlung, photoelectric effect, Compton scattering, pair production, exponential absorption, absorption coefficient and half-thickness. The film concludes with a section on the interpretation of composite absorption curves, such as are obtained when counting samples with beta rays having two energy maxima, beta and gamma, or positron and annihilation radiation. Released by the Armed Forces Institute of Pathology.

7. Atomic Theory, Part I, Atomic Physics Series - 10 minutes; 16 mm; black and white; sound.

This film develops the rebirth of the atomic theory of matter beginning with the discovery of the law of definite proportions and Dalton's basic atomic theory. Mendeleeff's periodic table, Brownian movement, and the early experiments of Michael Faraday, showing that electric current must be composed of unit particles of electricity, are reviewed. The film concludes with a summary by Lord Rutherford. Released by United World Films, Inc.

8. Rays from Atoms, Part II, Atomic Physics Series - 10 minutes; 16 mm; black and white; sound.

This film reviews the early investigations of cathode rays showing that they are small particles which possess mass and charge and which travel in straight lines. Included are the basic principles of the experiments by J. J. Thompson. The film concludes with a review of the nature of positive rays and the discovery of x-rays by Roentgen. Released by United World Films, Inc.

9. Nuclear Structure, Part III, Atomic Physics Series - 19 minutes; 16 mm; black and white; sound.

This film begins with the discovery of natural radioactivity by Henri Becquerel in 1896, showing the effect of radiation on a photographic plate. This is followed by a review of the work of the Curies in isolating radium, the various types of radiation and their behavior in a magnetic field, and Rutherford's experiments showing that alpha particles are helium nuclei and how the pattern of their scatter by a gold foil served as a basis for the classical concept of the atom. The structure of selected simple atoms is then shown and the importance of the atomic number of elements, rather than the atomic weight, is developed through a review of the results of Moseley's experiments with characteristic x-rays. The film concludes by showing the realization of the dream of the alchemist to change one element into another, when Rutherford bombarded atoms of nitrogen with alpha particles, forming oxygen and hydrogen. Released by United World Films, Inc.

RADIATION HAZARDS: ASSIGNMENT AND PROTECTION

10. Introduction to Radiation Detection Instruments - 19 minutes; 16 mm; black and white.

This film presents the fundamentals necessary to an understanding of the theory of operation of radiation detection instruments. Emphasis is on the need for instruments for detection as our senses are unable to detect ionizing radiation. It includes a brief description of a dosimeter, pocket chamber, ion chamber survey meter, alpha portable survey meter, Geiger-Mueller tube and a Geiger-Mueller-type survey meter. A discussion is included of the penetrating ability of alpha, beta, and gamma rays, calibration of instruments, and their use in the event of disaster. Laboratory instruments are shown as used in more detailed analysis. A brief description of film-badge monitoring is also included. A radiological monitoring group is shown in action as they evaluate various radiological hazards. Released by the Armed Forces Special Weapons Project.

11. Practical Procedures of Measurement (The Radioisotope - Film III) - 48 minutes; 16 mm; black and white.

The user of radionuclides must be concerned with radiation measurements for safety and experimental purposes. Several sequences cover the principles and use of various types of instrumentation, with emphasis on the Geiger counter, and illustrate background, threshold value, plateau, and counting statistics. The remainder of the film is devoted to absolute measurement, wherein the true activity of a sample is determined by using a calibrated standard, and comparative measurement, wherein the activity of a sample is compared with that of a control sample in order to determine the quantitative change in activity which resulted from decay or dilution. Released by the Armed Forces Institute of Pathology.

12. Practice of Radiological Safety - 33 minutes; 16 mm; black and white.

Depicts visit through a radioisotope laboratory, and discusses handling of radioisotope shipments; preparation of therapeutic doses; need for and function of a local radioisotope committee; laboratory design; decontamination; use of shielding; measurement of personnel exposure, and other topics pertinent to health safety. Released by the Army Surgeon-General.

13. Principles of Radiological Safety: I. Roentgen Unit, and II. Internal Radiation Dose. - 51 minutes total; 16 mm; black and white; sound.

Introduced in these two films are concepts of internal and external, acute and chronic, radiation exposure, illustrated by a historical sequence on hazards associated with x-ray and radium therapy and radium dial painting. Ionization from external and internal alpha, beta, and gamma radiation is studied, with detailed explanations of roentgen and "equivalent" or "energy" roentgen units. Maximum permissible exposure and the theory of radiation-measuring instruments are discussed. Formulae are developed for computing dosage rates from internal sources. Concepts of single and continued uptake, physical decay, and biological elimination of activity, biological half-life, and effective half-life are considered. Emphasis is on the responsibility of the radionuclide user to other personnel of the laboratory and to the public. Released by the Army Surgeon-General.

14. The Roentgen - 15 minutes; 16 mm; black and white; sound.

This film develops the concept, definition, and measurement of the Roentgen unit. By way of introduction, the relative penetrating and ionizing abilities of the various types of ionizing radiation are compared. This is followed by illustrations of the production of ion pairs by x and gamma radiation and a pictorial demonstration of the concepts of secondary electron equilibrium and air equivalent walls.

15. Human Radioactivity Measurements - 9 minutes; 16 mm; color and sound.

This film shows a method developed at Los Alamos Scientific Laboratory to monitor personnel exposed to possible intake of gamma-emitting materials, and to study the retention and excretion of radionuclides by the body. The liquid scintillation counter is large enough to contain a man and sensitive enough to detect even the minute amounts of his natural gamma radioactivity. Released by the U.S. Atomic Energy Commission.

16. Ionizing Radiation in Humans - 15 minutes; 16 mm; color and sound.

Central in this reel is Argonne National Laboratory's whole-body counter, its design and operation, its use in determining identification, quantity, and location of internally deposited radionuclides. The tilting chair, one-meter arc, techniques in accumulation of data, and collimating of the crystal are among subjects of footage. Released by the U.S. Atomic Energy Commission.

17. Living with Radiation - 28 minutes; 16 mm; color and sound.

A semi-technical documentary film discussing in detail the radiation-safety program of the national atomic energy program, using the procedures at AEC's National Reactor Testing Station in Idaho as the typical illustrative example. The film covers the separation-distance factor; storage and/or dispersal of radioactive wastes; protection of populations, water, crops, and livestock by air monitoring and environmental monitoring; and protection of workers by film badges, special clothing, radiation counters, shielding, remote control devices, decontamination procedures, and biochemical studies. Released by the U.S. Atomic Energy Commission.

18. Offsite Monitoring of Fallout During Nuclear Tests - 29 minutes; 16 mm; color.

This film describes the radiological safety activities of the Public Health Service in the offsite area of the U.S. Atomic Energy Commission's Nevada Test Site - a 300-mile radius, including portions of Nevada, California, Utah, and Arizona. Included is a description of the training provided PHS Commissioned Reservists from state health departments, universities, and industry; monitoring and public information activities of representative PHS Zone Commanders; methods of collection, and laboratory analysis of environmental samples. Produced for and by the U.S. Public Health Service.

19. A Matter of Contamination Sense - 10 minutes; 16 mm; black and white.

This is a short, relatively humorous film showing a case of contamination in a nuclear installation. It stresses common sense and points out several possible ways a person might become unknowingly contaminated. Produced by G. Buckland Smith in association with the Film Producers Guild for the United Kingdom Atomic Energy Authority.

20. Criticality - 22 minutes; 16 mm; color.

This film discusses the theory of criticality, and describes the conditions which will produce it, emphasizing the importance of mass, shape and moderation. Included are many examples of the precautions which must be taken in industry to assure that fissionable material will not become critical. Produced by G. Buckland Smith in association with the Film Producers Guild for the United Kingdom Atomic Energy Authority.

BIOLOGY, MEDICINE: CLINIC AND LABORATORY

21. Radiation: Physician and Patient - 45 minutes; 16 mm; color.

This film is essentially an informal talk by Dr. Richard H. Chamberlain (of the University of Pennsylvania School of Medicine and member of the National Committee on Radiation Protection and Measurements) about medical radiology - the problem it raises; its biological effects; its physical behavior; and its proper use in clinical examination. Many eminent authorities in the radiobiological field, who are interviewed in their own laboratories, discuss such matters as radiation effects on chromosomes, somatic effects of radiation, and dosage problems in radiation and clinical applications. Produced by the American College of Radiology in cooperation with the Public

Health Service, supported by a grant from the Rockefeller Foundation.

22. You Can Be Safe From X-Rays - 10 minutes; 16 mm; black and white.

This film is intended for those whose occupations necessitate frequent exposure to x-radiations. Emphasized are the facts that penetrating radiation cannot be detected by one's five senses and radiation damage is cumulative and may not be noticed until permanent harm has been done. With illustrations in animation, the film is primarily designed for photofluorographic operators making mass chest x-ray surveys. It has many well-illustrated public health principles equally of interest to anyone engaged in x-ray work. Released by United World Films, Inc.

23. Teletherapy and Brachytherapy - 18 minutes; 16 mm; color and sound.

Diagnostic and therapeutic applications are shown for radionucludes such as cobalt-60, cesium-137, europium-152, iodine-131, and yttrium-90. Released by the U.S. Atomic Energy Commission.

24. Iodine-131 - 15 minutes; 16 mm; color and sound.

Diagnostic and therapeutic uses of the radionuclide iodine-131 are demonstrated for hyperthyroidism, thyroid cancer, and heart disease. Characteristics, techniques, and results are shown. Released by the U.S. Atomic Energy Commission.

25. Radiation as a Cause of Cancer - 20 minutes; 16 mm; color.

This film presents an explanation of the histological step-by-step formation of cancer in various experimental animals and man after exposure to ionizing radiations. A brief review is made of basic atomic structure, radioactivity, and interaction of radiation with tissue; free radical formation is discussed in some detail. Also shown is an interesting experiment in which rats are fitted with jackets lined with strontium-90 beads. After a suitable exposure and ensuing latent period, skin cancer appears in the test animals. The carcinogenic hazards of external skin exposure are compared with internal exposures resulting from the uptake of radioactive materials by the gastrointestinal tract and the respiratory system. Produced by Encyclopedia Britannica Films.

26. Modification of Radiation Injury in Mice - 10 minutes; 16 mm; color and sound.

Shows the effects of chemical protection on mice before radiation, and of bone marrow transplant after radiation exposure. Possible implications in treatment of some human diseases are discussed. Released by the U.S. Atomic Energy Commission.

27. Chromosome Labeling by Tritium - 15 minutes; 16 mm; color and sound.

A presentation of the advantages of tritium over other radionuclides as labeling material in autoradiography. Released by the U.S. Atomic Energy Commission.

RADIATION PUT TO WORK: NON-MEDICAL USES

28. The Atom Comes to Town - 29 minutes; 16 mm; color and sound.

A survey of the peacetime uses of atomic energy. Included: How heat from nuclear fission in a reactor can be used to make electricity, and what nuclear power will mean to the man on the street and to all of America: Scenes of various experimental and prototype power plants, with discussion of types, kilowatt capacity, principles and future developments: Explanation of radioisotopes - how they are made and how they are used to alleviate suffering and raise the standard of living; Uses of radioactive materials in diagnosis and treatment of disease: Use of radionuclides in agriculture for production of better crops: Atomic energy as a means of quality control in manufacturing and industrial operations (studies on washing machines, engine wear, tires, toothpaste, plastics): Food preservation by radiological techniques. Chamber of Commerce of the United States production released by the U.S. Atomic Energy Commission.

29. Industrial Applications of Radioisotopes - 57 minutes; 16 mm; Color.

This semi-technical film surveys the current widespread uses of radionuclides throughout American industry. Three major areas of use are described: nuclear gaging (thickness, density and level), radiography and tracing. Luminescence, static elimination, isotopic power and uses of high intensity radiation are covered briefly. Basic principles are explained by animation, followed by examples of in-plant uses. The film is designed to acquaint industrial management with the versatility, economy and ease with which radioisotope techniques can be adapted to plant requirements.

30. X-Ray Inspection - 21 minutes; 16 mm; black and white.

The theory of x-ray production and the application of the principle of differential absorption of x-rays in a medium to radiographic inspection of metallic artifacts are briefly discussed. The procedures of a simple radiographic inspection of a cast gear blank are shown in detail, from the technician's viewpoint. Some x-ray protection principles are mentioned. Released by United World Films, Inc.

31. Gauging Thickness with Radioisotopes - 4 1/2 minutes; 16 mm; black and white; sound.

This brief film explains how beta gauges are used for precise measurement and control of feedback apparatus in steel, plastics, rubber, and paper manufacturing. Released by the U.S. Atomic Energy Commission.

32. Methodology (The Radioisotope - Film IV) - 40 minutes; 16 mm; black and white.

Opening with a historical sequence showing the early work of Hevesy in studying plant metabolism using naturally occurring radionuclides, this film goes on to essential criteria to be considered for setting up tracer experiments. They include radiochemical purity, single chemical state, elimination of exchange error, knowledge of the degree to which the labeled molecules remain intact, avoidance of chemical effects, and avoidance of radiation effects. Also illustrated are the relative importance of economy of time and materials and the degree of accuracy that can be achieved when utilizing radionuclides in experimental work. Released by the Armed Forces Institute of Pathology.

The Problem of Radionuclide Analysis

I. INTRODUCTION

Those agencies responsible for conducting surveillance programs and evaluating environmental radiological health problems recognize the increasing need for improved methods in the determination of radionuclide concentrations in environmental samples (air, food, milk, water, vegetation). Such radionuclides are present throughout the environment and originate from a number of natural and man-made sources. For the purpose of this discussion, we will be concerned with the problem of analyzing the radionuclides present in environmental samples, regardless of origin.

There are two general methods of radioactivity analysis used in environmental surveillance, namely, gross and specific. Gross analysis consists of concentrating a large volume or weight of sample into a small, dense portion which is counted without regard to the radionuclide makeup of the sample. Specific analyses may be performed instrumentally by gamma counting the original sample or by performing radiochemical separation on the sample and then counting the purified, separated radionuclide fraction by alpha, beta, or gamma counting techniques.

II. HISTORICAL PERSPECTIVE

The development of nuclear energy received its first real impetus in 1942 when Fermi and his associates achieved a sustained and controlled nuclear fission chain reaction in a graphite uranium pile. During the period after 1942, extraction and separation processes were utilized on a large scale to obtain nuclear fuels for chain reacting systems, namely, the atomic bomb and nuclear reactors.

Accordingly, the early radionuclide assays were associated with nuclear fuel technology. Such assays were designed for analysis of fuel materials, induced radionuclides, and fission products of relatively high activities.(1)

They are still widely used in national laboratories and reactor facilities where it is common to analyze high activity samples. In contrast, environmental assays are designed for determining low-levels of radioactivity in samples. The analytical procedures involved in environmental assays are complicated by the need for quantitative recovery of added carriers and complete decontamination from other radionuclides.

A. Problems of Environmental Radionuclide Analysis

Due to the low levels of activity found in environmental samples, the volume or weight of sample taken for analysis is invariably quite large, on the order of liters or kilograms. The natural radioactivity which is ever present in environmental samples was of no concern to the earlier investigators who were analyzing "artificial" samples containing only fission products, induced activities, and actinide or trans-uranium elements. However, natural radioactivity is a very real problem in environmental samples, and analytical procedures must be devised to decontaminate from such radionuclides as K^{40}, Ra^{226}, and Th^{232}. Since the radioactivity concentrations of environmental samples are relatively low, no contamination from other radionuclides can be tolerated, and the carrier recoveries must be as nearly complete as possible.

B. Advantages of Gamma Spectral Analysis

While it is possible to devise schemes of radiochemical analysis for each element, it is not possible to separate nuclides of the same element by chemical analysis. The conventional instrumental techniques (2, 3) for identifying mixtures of beta emitting radionuclides by energy and half-life are useful if sufficient activity is present, the beta energies are widely separated, and the half-lives are quite different. Such a convenient situation is not found in practice when analyzing environmental samples.

It is fortunate, from an analytical standpoint, that very few radionuclides of public health concern are pure beta emitters. Since virtually all radionuclides emit gamma rays, as well as beta particles and/or alpha particles, an instrumental procedure of analysis utilizing the property of the discrete energy of gamma rays for identification, would seem to offer a feasible way of unscrambling complex radionuclide mixtures. An examination of radionuclide decay schemes, on the basis of photon energies, reveals that gamma ray analysis is feasible both for isotopic separation of radionuclides of the same element and radionuclide mixtures of different elements.

C. The Advent of Gamma Scintillation Spectroscopy

As early as 1899, Becquerel first noticed that scintillation or luminescence could be observed when radiation interacted with such substances as zinc sulfide, barium platinocyanide, and diamond.(4) Other observers, at about the same time, noted that it was possible to observe this luminescence under a microscope and to delineate the luminescence as a series of individual light flashes, rather than a steady flow. Rutherford and Geiger used this principle to count the individual light flashes produced by a radium C source on a zinc sulfide screen. The number of light flashes they observed was approximately equal to the number of counts they obtained on an electrical counter, thus establishing the reliability of the scintillation method of counting alpha particles.

Although the visual counting of scintillations was a very tedious, time consuming process, this was virtually the only method used for qualitative and quantitative analysis of alpha particles until after 1930.

During the 1930 period, there was an improvement in the development of electrical counters due to the advent of vacuum tubes and associated circuitry. The scintillation counting method was then virtually discarded until about 1947.

The reason for the "revival" of scintillation counting was the development of the photomultiplier tube.(5) This tube, which is an electron multiplier with a photoelectric cathode, allows a single light scintillation falling on the cathode to start a chain of events known as photoelectric multiplication. The resulting electrons produce a current which can be measured and registered as resulting from one pulse. In other words, what the early investigators were obliged to do visually could now be done electronically. The development of the photomultiplier tube was the real break-through in the development of scintillation spectroscopy, which is now one of the most versatile analytical tools available to the nuclear scientist. Concurrent with the development of the photomultiplier tube was the development of crystal detectors (both organic and inorganic), organic liquid detectors, and gaseous detectors, all of which are capable of producing scintillations when a particular type of radiation interacts with them. In the case of gamma radiation, the development of solid inorganic crystals for producing scintillations and the development of the photomultiplier tube for converting this light to current, were the two factors which made the gamma scintillation spectrometer possible. The time period from late 1940's to the present has seen many refinements of the early scintillation detectors.

III. CHEMICAL RADIONUCLIDE ANALYSIS VERSUS GAMMA SPECTRAL ANALYSIS

The radionuclide composition of environmental samples is quite variable and often complex. If each radionuclide in the sample is to be determined by radiochemical procedures which ultimately result in the purified fractions being beta counted, then very lengthy, costly, and complicated sequential radiochemical separations are involved. Also, very exacting sample preparation procedures are required before the samples can be beta counted.

A good example of the problem is milk. Experience has shown that the principal radionuclies in milk are the following(6)

1. Radioiodine - usually only I^{131}
2. Radiocesium - usually only Cs^{137}
3. Radiobarium - usually only Ba^{140}-La^{140}
4. Radiostrontium - usually only Sr^{89}, Sr^{90}-Y^{90}
5. Potassium - 40

While it is true that a variety of chemical methods for the radioassay of milk are available, a conservative estimate of time necessary for one experienced radiochemist to analyze one sample of milk would be a minimum of two days (this excludes the specialized case of radiostrontium which requires more time for ingrowth) for the chemistry and possibly one more day for the counting. The same milk sample could be counted directly on a gamma spectrometer in a period of two hours or less, and activity concentrations for I^{131}, Ba^{140}-La^{140}, Cs^{137}, and K^{40} determined in a matter of minutes. It should be pointed out, however, that this is an estimate of time for the "ideal" case and is not representative for other environmental samples where the radionuclide composition is not known.

IV. LIMITATIONS OF GAMMA SPECTROSCOPY

A. Radiostrontium

In the example mentioned above, it is obvious that Sr^{89} and Sr^{90}-Y^{90} (pure beta emitters) cannot be determined by gamma spectroscopy. This is a serious limitation since radistrontium is of great interest and concern to public health workers.

B. Cost

The expense of acquiring, installing, operating, and maintaining a gamma spectrometer is often prohibitive to those agencies operating on limited budgets. Although the expense can be justified on a long term basis, the initial outlay is sometimes difficult to absorb.

C. Level of Detectability

The minimum level of detectability for beta counting is determined by the efficiency of the counting instrument, the background of the counting instrument, the length of time the sample is counted, and, of course, the amount of activity in the sample above background.(7) Determination of activity concentrations of one (1) pc/liter of I^{131} by beta counting is quite feasible and lower concentrations could be reported and be statistically sound. Unfortunately, this is not the case, for instance, in the gamma spectral analysis of milk. Due to limitations in detection efficiency, instrument stability, and spectral complexities, the lower limits of detection are usually about 10 pc/liter for I^{131} or a factor of ten higher than for beta counting.

V. SUMMARY

Radionuclide analysis of environmental samples is a complex task. If the samples are to be analyzed radiochemically, a thorough knowledge of the chemistry of all the stable and radioactive interferences is necessary. If the samples are to be analyzed instrumentally, then a thorough knowledge of the operation capability and limitations of the instrumentation is necessary.

The gamma scintillation spectrometer is a powerful tool in the field of environmental radionuclide analysis, within certain limitations. While it by no means completely replaces wet chemical procedures or eliminates sample preparation, it results in saving of time in most cases and permits rapid identification of radionuclides in a variety of samples.

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GPO 881-060

Elements of Gamma Spectroscopy

I. INTRODUCTION

Gamma spectroscopy is the science (and sometimes art) dealing with the use of the gamma spectrometer and the subsequent analysis of the data obtained from the spectrometer.

A gamma ray spectrometer is a combination of a scintillation detector which will produce electrical pulses representative of the gamma photons incident upon the detector, and a pulse height analyzer which is capable of distinguishing electrical pulses of different magnitude and determining the number of pulses occurring at each magnitude. The data which is obtained from the spectrometer is in two dimensional form usually expressed as count rate versus energy.

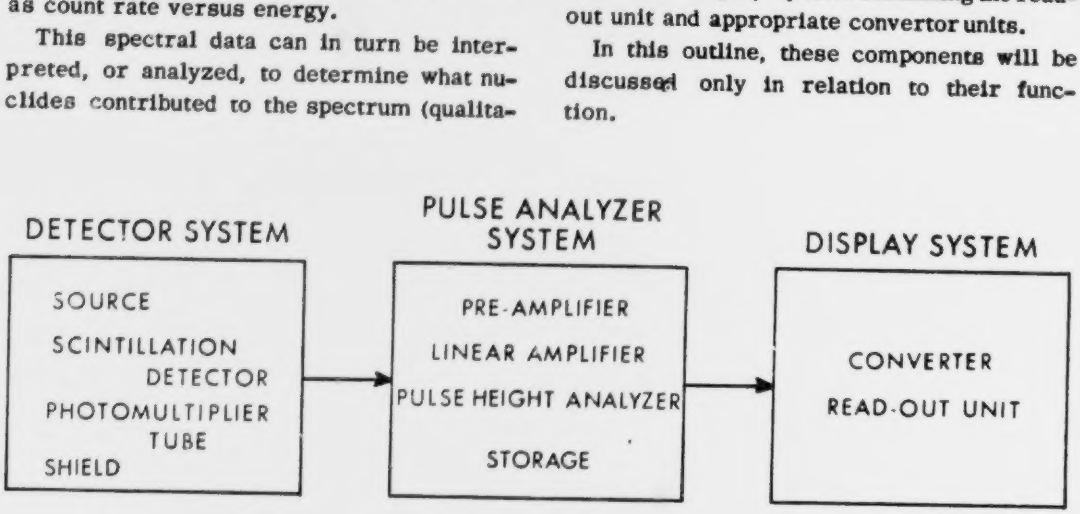
This spectral data can in turn be interpreted, or analyzed, to determine what nuclides contributed to the spectrum (qualitative analysis) and how much activity of each component nuclide there was (quantitative analysis).

II. COMPONENTS OF THE GAMMA SPECTROMETER

The entire gamma spectrometer can be broken down into three systems as indicated in Figure 1.

1. The detector system composed of a scintillation detector, photomultiplier tube, and shield.
2. The pulse analyzer system composed of the preamplifier, linear amplifier, pulse height analyzer, and storage unit.
3. The display system containing the read-out unit and appropriate convertor units.

In this outline, these components will be discussed only in relation to their function.


DETECTOR SYSTEM
SOURCE
SCINTILLATION DETECTOR
PHOTOMULTIPLIER TUBE
SHIELD
PULSE ANALYZER SYSTEM
PRE-AMPLIFIER
LINEAR AMPLIFIER
PULSE HEIGHT ANALYZER
STORAGE
DISPLAY SYSTEM
CONVERTER
READ-OUT UNIT

GAMMA SPECTROMETER COMPONENTS

Figure 1

A. The Detector System (See Figure 2)

1. SCINTILLATION DETECTOR

The function of the scintillation detector is to convert the energy of incident gamma photons to light energy. The amount, or intensity, of light produced is directly proportional to the energy transferred to the crystal when a gamma photon interacts with it. For each gamma interaction, there is produced one flash of light which is representative of that interaction.

2. PHOTOMULTIPLIER TUBE

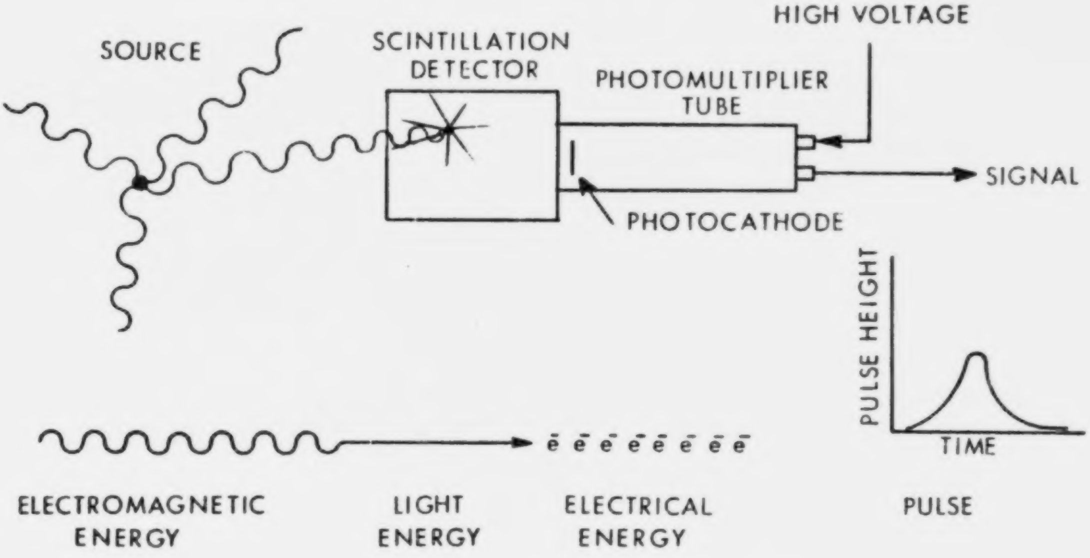
The light energy produced in the crystal is converted into electrical energy by means of the photomultiplier tube which is composed of the photocathode and the electron multiplier. The photocathode possesses the property that when it is struck by light, electrons are emitted from it in proportion to the amount of light incident upon it. The small number of electrons which are produced are then multiplied by accelerating them across a series of voltage potentials. The group of electrons which are produced from one gamma interaction form a pulse whose height is proportional to the energy transferred in the original interaction.

3. SHIELD

The purpose of the shield is to reduce the instrument background so that the effect of statistical variation in the net count rate will be as negligible as possible. The shield will also affect the shape of the net spectrum because of scattering of photons back into the scintillation detector.

B. The Pulse Analyzer System (See Figure 3)

In Getger or proportional counting systems, the individual pulses from the detector are simply recorded on a scaler to indicate


HIGH VOLTAGE
SOURCE
SCINTILLATION DETECTOR
PHOTOMULTIPLIER TUBE
SIGNAL
PHOTOCATHODE
PULSE HEIGHT
TIME
e e e e e e e e
ELECTROMAGNETIC ENERGY
LIGHT ENERGY
ELECTRICAL ENERGY
PULSE

THE DETECTOR SYSTEM

Figure 2

the total number of pulses, or counts, that have occurred in a given time period. However, the gamma spectrometer not only determines the number of pulses that have occurred but also classifies them into given energy ranges. This classification is done by the pulse analyzer system which contains a preamplifier, linear amplifier, pulse height analyzer, and storage unit.

1. PREAMPLIFIER AND LINEAR AMPLIFIER

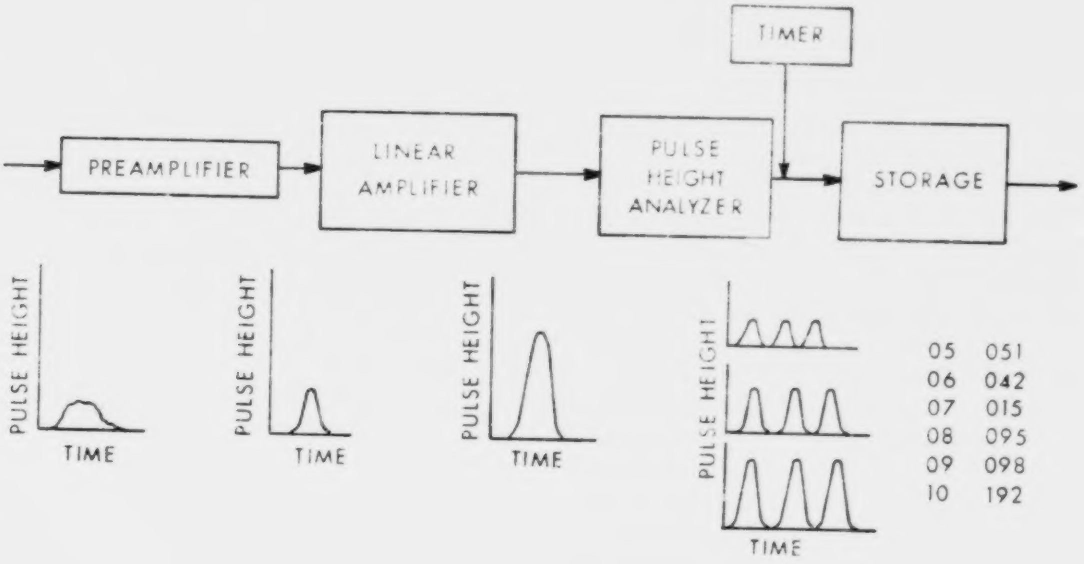
The function of these two units is to convert the weak, low amplitude pulse which is emitted from the photomultiplier tube to a stronger, higher amplitude pulse that can be readily handled by the succeeding pulse height analyzer. The two primary characteristics of these units are that they produce pulses of uniform shape and that the size of the pulse is in direct linear proportion to the pulse from the photomultiplier tube.

2. PULSE HEIGHT ANALYZER

The pulse from the linear amplifier is then fed into a pulse height analyzer which is capable of distinguishing pulses of different magnitude. The number of pulses of each magnitude is then recorded or stored by one means or another.

3. STORAGE

The storage unit is the filing system of the spectrometer. The information that is continuously generated in the pulse height analyzer is added together and compiled as a running total of the number of pulses in each pulse height range. At the end of the sample counting period, the information in the storage unit is a compilation of all the different pulses originating from the detector system during the counting period.


TIMER
PREAMPLIFIER
LINEAR AMPLIFIER
PULSE HEIGHT ANALYZER
STORAGE
PULSE HEIGHT
TIME
PULSE HEIGHT
TIME
PULSE HEIGHT
TIME
PULSE HEIGHT
TIME
05 051
06 042
07 015
08 095
09 098
10 192

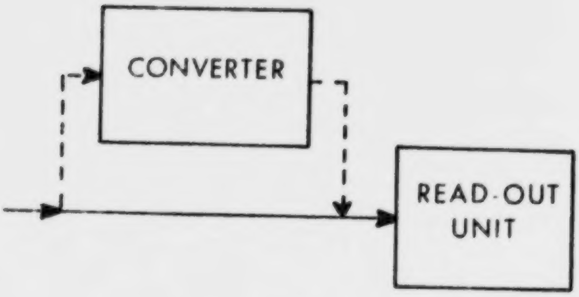
THE PULSE ANALYZER SYSTEM

Figure 3

C. Display System (See Figure 4)

1. READ-OUT UNIT

In single-channel and the earlier multichannel (voltage discriminator type) analyzers, the storage and read-out units are one in the same; mainly, a scaler or set of scalers. However, in the present computer type multichannel spectrometer, the information in the storage unit is in a form that cannot be visually interpreted and must be transformed to a physical or numerical representative of the data.



THE DISPLAY SYSTEM

Figure 4

2. CONVERTER UNITS

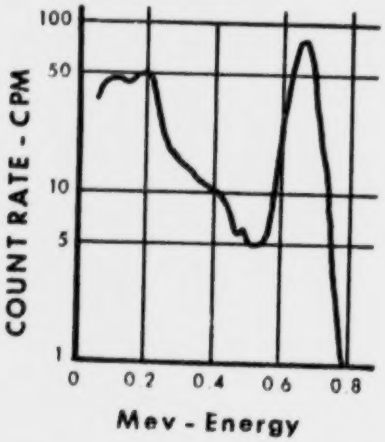
Many times the electrical form of the data coming from the storage unit is not compatible with the read-out unit and it must be changed. This compatibility can be achieved by use of separate converter units.

III. THE GAMMA RAY SPECTRUM

Gamma rays, emitted as a result of nuclear disintegrations, are monoenergetic and are characteristic of the nuclide which underwent disintegration; however, a given energy gamma ray is not necessarily unique to a particular nuclide. Also, most nuclides emit more than one gamma ray, thus adding to the complexity of the resulting spectrum.

A. Display of Spectra

The information which is fed out of the spectrometer is normally expressed in terms of two parameters, count rate and energy. Figure 5 shows a typical plot of a cesium-137 spectrum using these terms.



CESIUM-137 GAMMA SPECTRUM

Figure 5

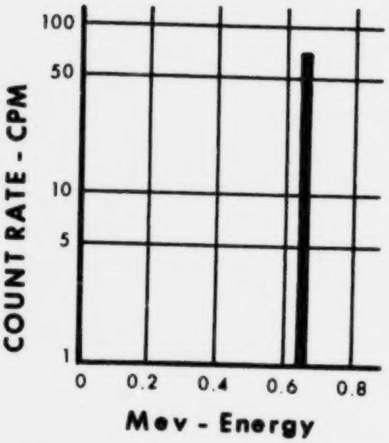
1. "IDEAL" SPECTRA

As gamma rays are monoenergetic, it would be expected that a "line spectrum" such as shown in Figure 6 would be formed rather than the type shown in Figure 5. The difference between the two spectra is due to the various mechanisms of gamma interaction and to the inherent limitations of the gamma spectrometer.

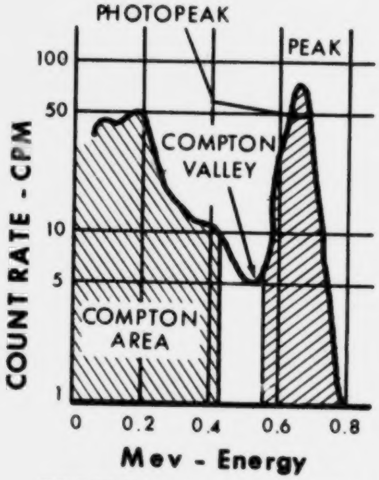
2. ACTUAL SPECTRA

It is useful to describe the actual spectrum that is obtained from a spectrometer by means of several terms which refer to the prominent portions of the spectrum. Figure 7 illustrates these terms.

a. ~~Photopeak~~ Peak - the energy at a spectral peak which represents the energy of an interacting gamma photon.



"IDEAL" LINE SPECTRUM OF CESIUM-137
Figure 6



DESCRIPTIVE TERMS OF
GAMMA SPECTRA

Figure 7

b. Photopeak - the area under a spectral peak, expressed as counts per minute, which is due to a complete energy transfer of an initial gamma photon.

c. Compton area - the area under the portion of the spectrum which is due to partial energy transfer by Compton interaction.

d. Compton valley - the low portion of the spectrum immediately before the photopeak which is due to the absence of Compton photons in this energy range.

B. Relationship of Peak Height to Activity

A fundamental property of gamma spectra is that the relative shape (the proportion of any portion of the spectrum to any other portion) is constant. This principle is illustrated in Figure 8 which shows that if the activity of the sample is doubled, the count rate of each portion of the curve is also doubled.

Important corollaries to the "constant shape principle" are that the activity of a sample is proportional to:

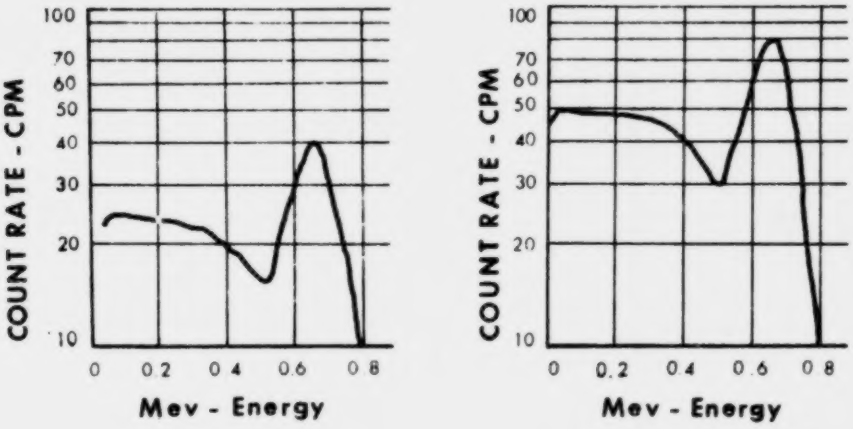
1. The total count rate under the entire spectrum
2. The count rate of the photopeak channel
3. The count rate in the photopeak area
4. The count rate of any portion of the spectrum

C. Composite Spectrum

A second fundamental principle of a gamma spectrum is that the composite spectrum of two or more nuclides is equal to the sum of the spectra of each component as shown in Figure 9.

V. SUMMARY

Gamma spectroscopy deals with both the gamma spectrometer and with the interpretation of the gamma spectrum. The overall function of the spectrometer is to provide a gamma spectrum which is representative of the gamma photons interacting with the scintillation detector. The actual spectrum differs from the ideal spectrum because of limitations of the spectrometer and the various possible mechanisms of gamma interaction.

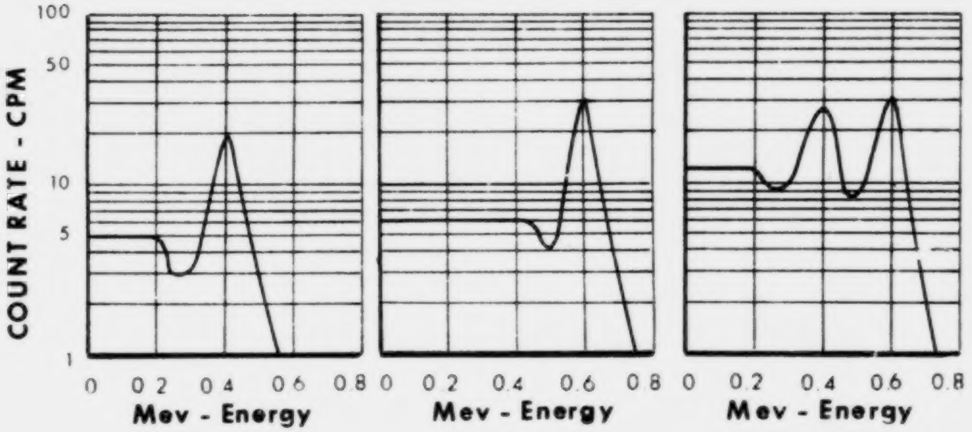


400 pc- Cs 137

800 pc - Cs 137

EFFECT OF ACTIVITY ON GAMMA SPECTRA

Figure 8



NUCLIDE X

NUCLIDE Y

NUCLIDE X+Y

COMPOSITE SPECTRA

Figure 9

REFERENCES

1. Crouthamel, C. E. "Applied Gamma-Ray Spectrometry". The MacMillan Company. New York, 1959.
2. Price, W. J. "Nuclear Radiation Detection". McGraw-Hill Series in Engineering, McGraw-Hill Book Company, Inc., New York. 1958.

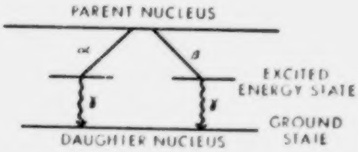
Interaction of Gamma Rays

I. FUNDAMENTALS

A. Introduction

Gamma rays, designated by the Greek symbol γ, are a type of electromagnetic radiation and move with the speed of light. They have zero rest mass and no net electrical charge. Gamma rays arise from atomic nuclei which are in an excited energy state. Since the atomic nucleus exists only in definite energy levels, gamma rays from a given radionuclide are emitted with discrete characteristic energies. These discrete energies may be termed "bundles" or "photons" of energy.

Gamma photons are usually emitted as an energy adjustment of the atomic nucleus following an alpha (α) or beta (β) emission. Schematically, the process can be pictured in Figure 1.



DECAY SCHEME

Figure 1

Gamma photons can also be emitted as a straight energy transition - without the immediate prior emission of an alpha or beta particle, such as the case of strontium-85.

Several different energy gamma rays may arise from any one parent nucleus, depending upon the variableness of prior alpha or beta energies. Some radionuclides may emit as many as five or more different energy gamma rays in association with their decay to a ground state; e.g., I^{131} emits five different energy gamma rays in its decay to stable Xe^{131}.

The energies of the gamma rays emitted from any given radionuclide are characteristic of the nuclide; however, they are not unique. Several different radionuclides may emit gamma rays of the same energy; thus creating difficulties in gamma spectral analysis.

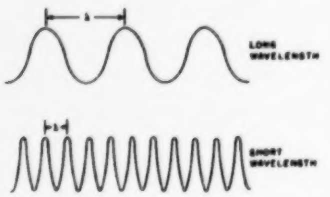
B. Properties of Gamma and X-rays

Gamma rays are similar to x-rays in that both are electromagnetic radiations and travel with the speed of light. However, they do differ in their origins; x-rays originating from transitions between electronic energy levels (orbital electron shells), and gamma rays originating from transitions between nuclear energy levels (nucleus).

Since electromagnetic radiation creates wave-like disturbances and travels with the speed of light, the energy of a photon can be evaluated. The concept of waves and wavelength is perhaps best illustrated by comparison to the disturbance created if a stone is dropped vertically in the center of a pool of water. As the stone strikes the water, a series of ripples or waves will be seen to move outward in concentric circles. The series of crests and troughs formed constitute a wave motion as shown in Figure 2, the distance between any two successive crests or troughs is known as the wavelength, which is usually represented by the Greek letter λ (lambda).

Since all electromagnetic rays travel at the same speed, (30 billion cm/sec or 186,000 miles/sec), the number of waves (crests) passing a certain point (or the frequency with which the crests pass) will decrease with increasing distance between crests (wavelength). This quantity is called the frequency and is represented by the Greek letter ν (nu). The relation between the wavelength λ, the frequency ν, and the velocity of light (C) is given by the equation

$$\nu = \frac{C}{\lambda}$$



ELECTROMAGNETIC RADIATION

Figure 2

The energy of an electroomagnetic radiation[1] can be expressed by the equation,

E = h (1), where

E = energy in Mev

h = Planck's constant = 4.14×10^{-21} Mev - Sec.

ν = frequency (cycles/sec)

The frequency of x-rays varies from about 10^{18} to 10^{23} cycles/sec. The frequency of normal gamma rays varies from about 5×10^{18} to 10^{21} cycles/sec. Therefore; the energies of most x-rays usually vary from a few Kev up to several hundred Mev, and the energies of gamma rays from a few Kev to several Mev. Gamma rays and x-rays differ from radio waves and visible light waves only in having much shorter wavelengths.

C. Interaction of Gamma Radiation with Matter

All radiation possesses energy either inherently, as in the case of electromagnetic radiation, or as kinetic energy of motion in the case of the particulate radiations. Absorption of radiation is the process of transferring this energy to atoms of the medium through which the radiation is passing. To say that radiation interacts with matter is to say that it is either scattered or absorbed.

1. TYPES OF ENERGY TRANSFER

The transfer of energy from the emitted particle or photon to atoms of the absorbing material may occur by several mechanisms but, of the radiations commonly encountered, the following two are the most important:

a. Ionization

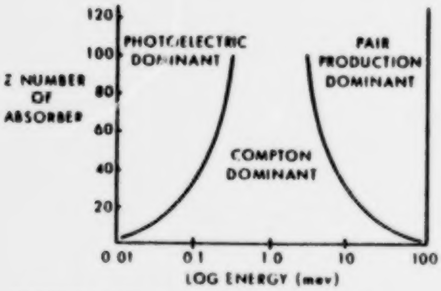
Any process which results in the removal of an electron (negative charge) from an atom or molecule thereby leaving the atom or molecule with a net positive charge is termed ionization.

b. Excitation

The addition of energy to an atomic or molecular system, thereby transferring it from its ground state to an excited state is termed excitation. Depending on the type of interaction, either the atomic nucleus or one of its orbital electrons may absorb the excitation energy.

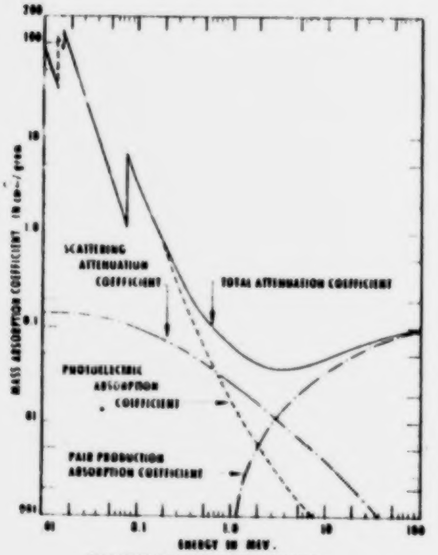
2. GAMMA PHOTON INTERACTION

Since gamma photons are basically only distortions in the electromagnetic field of space, they interact electrically with atoms to produce ionization, even though they possess no net electrical charge. Theoretically, there are twelve processes of gamma ray interaction with matter.[2] The three most important interactions are the photoelectric effect, the Compton effect, and pair production. The predominance of the particular interaction depends upon the gamma photon energy and the Z number of the absorbing medium (see Figures 3 and 4).



INTERACTION PREDOMINANCE

Figure 3



MASS ABSORPTION COEFFICIENTS FOR LEAD
CONTRIBUTION OF THE PHOTOELECTRIC EFFECT, COMPTON SCATTERING, AND PAIR PRODUCTION TO THE TOTAL ATTENUATION COEFFICIENT

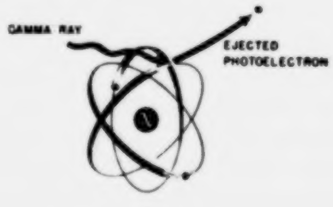
Figure 4

II. PHOTOELECTRIC EFFECT

A. Process

The photoelectric effect is predominant when conditions of low gamma ray energies and high atomic weight absorbers exist. The gamma wave, or photon, imparts all of its energy to an orbital electron of some atom; and, since the gamma photon consisted only of energy in the first place, the photon vanishes, thus the process can be considered as a complete energy loss process. In order for momentum to be conserved, another body must be present, so gamma photons can not interact with free electrons by the photoelectric effect.

The gamma photon energy is imparted to the orbital electron in the form of kinetic energy of motion, and this greatly increased energy overcomes the attractive force of the nucleus for the electron and causes the electron to fly from its orbit with considerable velocity (see Figure 5).



PHOTOELECTRIC EFFECT
(PRIMARILY LOW ENERGY PHOTON)

Figure 5

The kinetic energy of the ejected electron (called a photoelectron) can be expressed as
KE = Eg - P (2)

where KE = kinetic energy of photoelectron

Eg = energy of incident gamma photon

P = energy to remove electron

= binding energy of electron in atom

P is usually small in relation to Eg and can be neglected. When $Eg \ll M_0C^2$ (rest mass of electron = 0.51 Mev), the photoelectron flies off predominantly at right angles to the photon beam. At higher energies near the upper level of photoelectric interaction (0.1-1.0 Mev), the photoelectron flies off in the same direction of incident photon.(4)

When the photoelectron is ejected an ion-pair results. The photoelectron has sufficient energy to knock other electrons from the orbits of other atoms; thus producing secondary ion-pairs until all of its energy is expended.

B. Variables Affecting Process

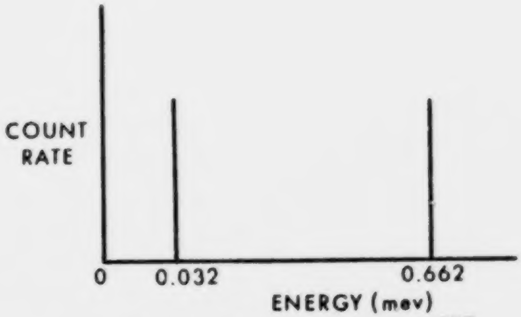
The probability of a gamma photon interacting by the photoelectric effect with a given orbital electron is dependent upon the orbital shell of the particular electron, the atomic number of the absorbing medium, and the photon energy. The more tightly an electron is bound in the Coulomb field of a nucleus, the more probable is photoelectric interaction. When the incident photon energy exceeds the K-shell binding energy, approximately 80% of the photoelectric interactions occur in the K-shell and the majority of the remaining 20% occur in the L-shell.(2) The photoelectric probability cross-section is approximately proportional to the fifth power of the atomic number of the absorber material in the incident photon energy (Eg) exceeds 0.1 Mev. When $Eg < 0.1$ Mev, the photoelectric probability cross-section is proportional to $Eg^{1/3}$; When Eg is between 0.1-0.5 Mev, the cross-section is proportional to $Eg^{1/2}$; and when $Eg > 0.5$ Mev, the cross section is proportional to $\frac{1}{Eg}$ (5).

C. Secondary Effect

After the photoelectron is ejected, and as the "empty space" in the electron shell is being filled with another electron from outer shell, the excess energy present can be emitted by one or more characteristic x-rays or an ejected outer orbital electron, known as an Auger electron.(2) As an x-ray is produced, it may either interact in the detector by the photoelectric effect or escape. Since the energy of the x-ray is much smaller than the energy of the incident gamma photon, the probability of its interaction by photoelectric effect is relatively greater.

D. "Ideal" Spectrum

If a spectrometer system was capable of discretely measuring each photoelectric occurrence from the gamma emissions of a single nuclide, and these emissions were of one energy and interacted only by photoelectric, the resultant "idealized" spectrum would be a "line" spectrum corresponding to the full energy of the gamma photon and the characteristic x-ray. For example; Cs^{137} would have a "line" peak at 0.662 Mev (gamma peak) and 0.032 Mev (x-ray peak), see Figure 6.



IDEAL SPECTRUM FOR Cs^{137}

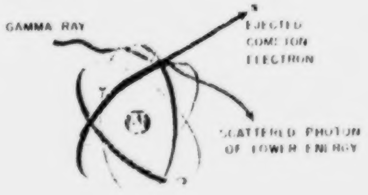
Figure 6

When gamma photons have energies much in excess of that required to eject an electron, they do not interact by photoelectric. The probability decreases as the incident photon energy increases; thus introducing another type of interaction.

III. COMPTON EFFECT

A. Process

The Compton Effect, the dominant effect at intermediate energy levels and low atomic weight absorber material (see Figure 3), provides a means of partial energy loss for the incoming gamma ray. The effect may be considered as an in-elastic, incoherent scattering of the rays by atomic electrons. Again the gamma ray appears to interact with an orbital electron of some atom, but in the case of Compton interactions, only a part of the energy is transferred to the electron, and the gamma ray "staggers on" in a weakened condition (see Figure 7). The high velocity electron, now referred to as a Compton electron, produces secondary ionization in the same manner as does the photoelectron, and the weakened gamma ray, called a scattered photon, continues on until it loses more energy in another Compton interaction or disappears completely via the photoelectric effect. The incoming photon may have several scatterings before its final escape or absorption.



COMPTON EFFECT
(PRIMARILY MEDIUM ENERGY PHOTON)

Figure 7

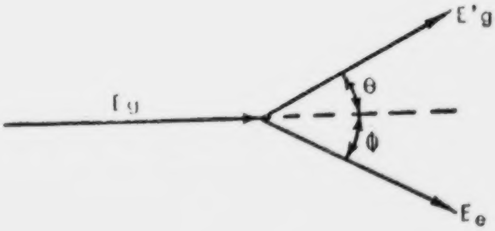
Compton scattering will generally involve the outer orbital electrons of an atom and does not produce a significant amount of K or L x-rays, except in lighter elements.(2)

Theoretically the Compton electron may have an energy value up to a maximum value (the Compton edge = Eg-Eg/1+4Eg). Since the Eg is large in comparison with the electron binding energy, the electron binding energy is neglected, and the remaining energy in the energy transfer is carried away by the scattered gamma photon. The variable energies of the Compton electrons give rise to the "Compton Continuum" in scintillation counting.

B. Energy Transfer

The actual loss of energy by Compton Effect depends on the angle between the direction of the scattered photon and the incident photon, this angle depending upon Eg.

Since momentum must be conserved the Compton electron must recoil at an appropriate angle. By knowing the angle between the scattered photon and incident photon, and the incident photon energy; the scattered photon energy and the Compton electron energy can be calculated from the law of conservation of momentum as follows.(5) See Figure 8.



COMPTON SCHEMATIC

Figure 8

$$E'g = \frac{Eg}{1 + \alpha\,(1 - \cos\theta\,)} \qquad (3)$$

$$Ee = Eg \left[1 - \left\{\frac{1}{1 + \alpha\,(1 - \cos\theta)}\right\}\right] \quad (4)$$

where Eg = incident photon energy

$E'g$ = scattered photon energy

Ee = Compton electron energy

θ = angle between direction of scattered photon and direction of incident photon

ϕ = angle between direction of Compton electron and direction of incident photon

$$\alpha = \frac{\text{incident photon energy}}{\text{energy equivalent of rest mass of electron}} = \frac{Eg}{Mc^2}$$

M = rest mass of electron

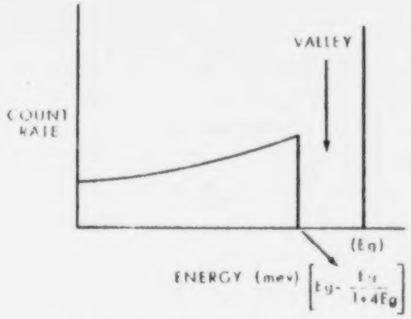
c = speed of electron

The Compton electron receives the least transfer of energy in a "grazing" collision in which the incident photon continues with its initial frequency in the forward direction, and the electron is ejected with very nearly zero velocity in a direction perpendicular to that of the incident photon path. The Compton electron receives the greatest transfer of energy in a completely elastic collision whereby $\theta = 180^\circ$, thus ejecting the Compton electron in a direction coinciding with the incident photon direction. In normal Compton interactions, the Compton electron flies off predominantly in the direction of the incident photon, the more nearly so the higher the fraction of photon energy it carries away and the higher the incident photon energy. When $Eg \ll Mc^2$ (0.51 Mev), Ee is negligible, when $Eg \gg Mc^2$, Ee approaches Eg as θ increases.(1)

The majority of Compton interactions of photons with $Eg > 50$ Kev will transfer less than 50% of the incident energy to the electron because there is a lower probability for $> 90^\circ$ scattering of the photon. "The average fraction of the incident photon energy lost per Compton interaction is about 14 per cent at 100 Kev, 34 per cent at 500 Kev, 44 per cent at 1.0 Mev, and 68 per cent at 10 Mev".(6) Gamma rays with energies < 50 Kev scatter forward and backward with almost equal probability.(5)

C. "Ideal" Spectrum

In the scintillation counting of Compton interactions the photomultiplier tube can "see" a spectrum of Compton electron energies due to the variableness of the energy transfer. The photomultiplier tube can also "see" several successive Compton events from one photon as one event because the interaction events occur in a much shorter time than the generation and decay time of the scintillation light pulse. Therefore, an idealized spectrum of one nuclide with $Eg > 50$ Kev would consist of a spectrum of activity from very low energies up to the Compton edge, no activity in the Compton valley, and a photopeak composed of a line spectrum with an energy of Eg. See Figure 9.



IDEAL COMPTON SPECTRUM

Figure 9

In actual spectra a pure line photopeak will not exist and the Compton valley will contain some counts due to the inherent statistics of the process.

IV. PAIR PRODUCTION

A. Process

Pair production, the third type of interaction, is much rarer than either the photoelectric or Compton Effect, predominating at high gamma photon energies and high atomic weight absorber materials. Pair production is impossible unless the gamma ray possesses at least 1.02 Mev (twice the rest mass of an electron) of energy. (Practically speaking, it does not become important until approximately 2 Mev).

1. PRODUCTION OF POSITRON AND ELECTRON

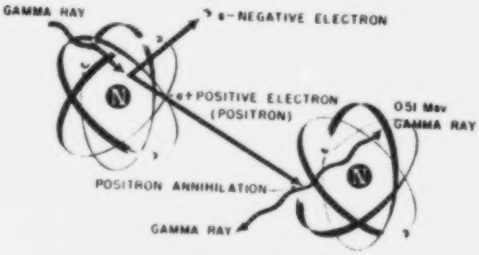
In pair production a gamma photon simply disappears in the vicinity of a nucleus, and in its place appears a pair of electrons - one negative, one positive (called a positron). The principal function of the nucleus is to allow conservation of momentum when the photon transfers its entire energy to the recoil particles. The negative and positive electrons are produced in order to conserve charge. The mass of these electrons has been created from the pure energy of the photon, according to the familiar Einstein equation $E = Mc^2$ where (E) is energy, in ergs, (M) is mass, in grams, and (c) is the velocity of light in cm/sec. If thre is any excess energy in the photon above the 1.02 Mev required to create two electron masses, it is simply shared between the two electrons as kinetic energy of motion, and they fly out of the atom with great velocity. Possibly slightly more than one half the available kinetic energy goes to the positron because it is repelled by the nucleus.(1)

In most instances the electron and positron are projected from the nucleus predominantly in the direction of the incident photon, especially when the photon energy, Eg, and hence its momentum, are very large. Most of the electrons and positrons are confined to directions within $1/\alpha$ radians from the photon direction

$$(1/\alpha = \frac{0.51\ \text{Mev}}{\text{Eg}}) \qquad (4).$$

2. INTERACTION OF POSITRON AND ELECTRON

The negative electron behaves in exactly the ordinary way, producing secondary ion-pairs until it loses all of its energy of motion. The positive electron also produces secondary ionization so long as it is in motion, but when it has lost its energy and slowed almost to a stop, it encounters a free (negative) electron somewhere in the material. The two are attracted by their opposite charges, and, upon contact, annihilate each other, converting the mass of each into pure energy. Thus two gammas of 0.51 Mev arise at the site of the annihilation. See Figure 10.



PAIR PRODUCTION
(HIGH ENERGY PHOTON, >102 Mev)

Figure 10

The life of positrons in liquids and solids are approximately 10^{-9} to 10^{-10} seconds.(7)

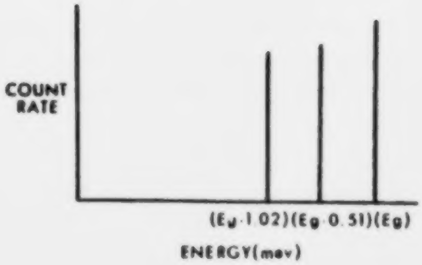
The two annihilation photons are emitted ~ 180° with respect to each other, but random with respect to the incident photon direction. The reason the annihilation photons are back is to conserve momentum. The ultimate fate of the annihilation gammas is either photoelectric absorption or Compton scattering followed by photoelectric absorption, or escape from the material.

B. Variables Affecting Process

The probability of pair production is dependent upon the gamma photon energy and the atomic number (nuclear charge) of the absorber material. The probability of pair production increases approximately linearly with photon energy starting from zero at the threshold energy, 1.02 Mev, and leveling off at higher energies. The probability for pair production approximately increases as the square of the atomic number of the absorber material. These probabilities explain why gamma photons from Tl^{208}, (Eg = 2.62 Mev), readily produce positron-electron pairs when they pass through lead.$^{(8)}$

C. "Ideal" Spectrum

An idealized spectrum for pair production would be quite complex due to the Compton scatter spectrum of the annihilation photons or lack of spectrum due to the escape of one or both anihilation photons. Peaks would be expected at energies of Eg, Eg-0.51 Mev, and Eg-1.02 Mev, see Figure 11. The spectrum can have energies ranging from Eg downward.



IDEAL PAIR PRODUCTION SPECTRUM

Figure 11

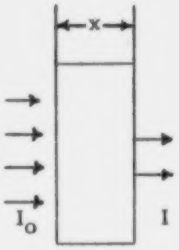
V. ABSORPTION OF GAMMA RADIATION

A. Process of Absorption

Since gamma rays are electromagnetic radiations, they are similar to visible light in that they suffer an approximately exponential attenuation (decrease is dose rate) upon passage through a semitransparent medium. That is to say, the ratio of the intensity of the beam emerging from an absorber to the intensity of the beam incident upon the absorber is equal to $e^{-\mu x}$, where (μ) is a constant depending upon the material with units reciprocal to those of (x), and (x) is the distance traveled by the beam in the material. Thus the equation can be written as:

$$I = I_0 e^{-\mu x} \qquad (5)$$

where I = intensity or count rate after passing through "x" distance of absorber material (See Figure 12).



ABSORPTION PROCESS

Figure 12

I_0 = intensity or count rate from a given source when no absorber is used.

μ = linear attenuation coefficient, units reciprocal to x units.

x = distance of concern in absorber material.

B. Good Geometry

The above relationship only holds true when the gamma beam in homogenous, straight, and collimated; and the absorber material is "thin". Such conditions are termed "good geometry".

Values of (μ), the linear attenuation coefficient, or of μ/p, the mass attenuation coefficient, may be found in references such as the Radiological Health Handbook,(9) or the Handbook of Chemistry and Physics.(10) The attenuation coefficient varies with the atomic number of the material, and with the energy of the gamma radiation involved.(11)

The linear attenuation coefficient is equal to the summation of the photoelectric absorption coefficient, the Compton attenuation coefficient, and the pair production attenuation coefficient.(12)

A useful quantity is the half-value layer, or half-thickness, which is the thickness of absorber required to reduce the intensity to one-half the initial value. The relationship between the linear attenuation coefficient, μ, and the half-value layer (half-thickness), $X_{\frac{1}{2}}$, can be derived from equation (5) above by substituting $X_{\frac{1}{2}}$ for x, and $\frac{I_o}{2}$ for I.

This can be simplified to give:

$$\mu \quad \frac{0.693}{X_{\frac{1}{2}}}$$

C. Poor Geometry

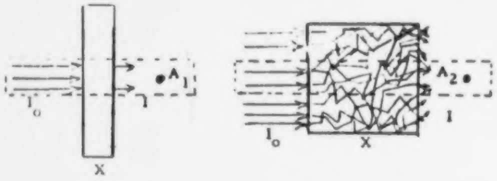
When conditions are such that several different energy gamma photons are present, either from one nuclide or several in an environmental situation, a strict exponential attenuation will not occur. The net effect of attenuation upon such mixtures of gammas is to filter out the weaker components first, leaving the harder components. Thus if one centimeter of lead were sufficient to reduce the initial beam intensity by one-half, more than one centimeter would be required to reduce the emerging beam intensity by one-half again, etc. Also in environmental situations the gamma rays are not collimated and thus are not incident upon the absorber material in a narrow beam. This "broad beam" or "poor geometry" situation results in considerable scatter or spread of the gamma rays, so that their final direction may be very different from their initial path.

Thus it may be seen from Figure 13 that the exposure at point A_2 is increased by the scattered radiation which it receives from the adjacent parts of the absorber material. The intensity at A_2 is observed to be higher than the $I = I_o e^{-\mu x}$ relation would predict, to allow for broad beam geometry a buildup factor "b" (always greater than unity) is inserted, and the basic equation becomes:

$$I = b\, I_o\, e^{-\mu x}. \qquad (6)$$

VI. SUMMARY

Gamma rays result from nuclear energy transitions. These gamma rays are electromagnetic radiation possessing discrete characteristic energies. They interact with



GOOD AND POOR GEOMETRY

Figure 13

matter by three major processes; photoelectric (complete energy transfer), Compton (partial energy transfer), and pair production (complete energy transfer with production of annihilation photons). The photoelectric effect dominates at low gamma energies and high atomic number absorber material, Compton at intermediate energies and low atomic number absorber material, and pair production at high energies and high atomic number absorber materials. The attenuation of gamma rays is dependent on the absorber material and the gamma energy.

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GPO 884-556

Characteristics of Gamma Spectra

I. INTRODUCTION

Before an adequate analysis of a gamma spectrum can be made, the general characteristics of a spectrum must be understood. A number of these characteristics have been mentioned in previous lectures in a different context. It is the purpose of this lecture to relate these characteristics to their use in identification of nuclides in a composite spectrum.

II. CHARACTERISTICS OF PHOTOPEAKS

Gamma-rays are characterized by monoenergetic emission. If the entire energy of the gamma photon is absorbed in the scintillation crystal, either in a single interaction or several successive interactions, a resulting peak (known as a photopeak) in the final spectrum is representative of the monoenergetic emission.

The energy of unknown photopeaks can be determined by knowing the energy calibration of the spectrometer which is expressed by the channel coefficient (Kev per channel).

III. CHARACTERISTICS OF COMPTON AREA

In the case of gamma photon interaction by the Compton effect, all of the incident gamma energy may not be absorbed by the crystal. The Compton interaction is characterized by the ejection of an electron and the scattering of the incident photon with a reduced energy. The scattered (Compton) photon may not undergo further interactions in the crystal and escape, leaving only the energy of the ejected electron to result in light emission in the crystal.

The Compton photons produced are not monoenergetic and result in a broad spectrum area, known as the Compton Continuum.

A. Compton Edge

The Compton Continuum has a definite upper energy limit referred to as the Compton edge. This occurs when the incident photon is scattered through an angle of 180°, resulting in the ejected electron receiving a maximum kinetic energy. This Compton edge energy may be calculated from the equation:

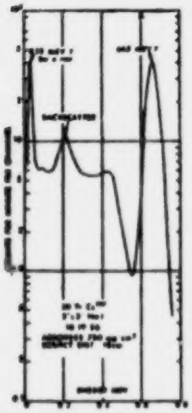
$$\text{Energy of Compton Edge} = Eg - \frac{Eg}{1 + 3.92\,Eg}$$

where Eg is the incident photon energy.

Many times, this edge may appear similar to a photopeak when distorted in a composite spectrum.

It should be noted that the Compton edge energy continually increases with increasing incident photon energy.

The Compton edge is more prominent in the spectrum of higher energy gamma rays because the probability cross section for the photoelectric effect, and also, the escape probability for scattered photons is larger. Examples of spectrums with a definite Compton edge are shown in Figures 1 and 7.



SPECTRUM OF CESIUM-137
Figure 1

B. Backscatter Peak

Another characteristic of the Compton area is the backscatter peak. When photons emitted from the sample interact by Compton effect with surrounding materials such as the background shield, the photons that are scattered through an angle of approximately 180° have a possibility of interacting with the crystal detector. Quite often these "backscattered" photons result in a peak which is superimposed on the Compton Continuum. The energy of the "backscatter peak" is equal to the energy of the incident gamma minus the energy of the ejected electron. This backscatter peak energy can be calculated by the equation:

$$\text{Energy of backscatter peak} = \frac{Eg}{1 + 3.92\,Eg} \quad (2)$$

where Eg is the incident photon energy.

The backscatter peak becomes less noticeable as the incident gamma energy increases. This is due to the fact that with increasing energy the total Compton cross section decreases, and the angular distribution of the scattered photons from Compton interaction becomes more predominant in the forward direction; therefore, the backscattering becomes less. Unlike the Compton edge, the backscatter peak has a maximum energy of 0.255 Mev.

The backscatter peak may also be affected by the geometry relationship between the sample and crystal. This effect is especially observed when comparing a well crystal with a flat crystal. The flat crystal will have a more prominent backscatter peak, due to the greater probability of the photons interacting with other substances before they interact with the crystal. Examples of backscatter peaks are shown in Figures 1 and 4.

C. Compton Ratio

In much of the work done in gamma spectroscopy it is advantageous to have a high ratio of interactions in the photopeak area (due to interactions where energy is totally absorbed) to interactions in the Compton area (due to loss of energy in Compton scattered photons). This ratio is known as the Compton ratio and is used to compare spectrometer systems. One method of increasing the Compton ratio is to enlarge the detector crystal. This increases the probability for the interaction of the Compton photons and the subsequent addition of their energy to the initial energy lost to the crystal. The result is that the overall interaction shows up in the photopeak of the spectrum.

IV. ESCAPE PEAKS

Escape peaks are denoted as spectrum peaks at some energy less than the photopeak energy, caused by the repetitious escaping of a discrete amount of energy from the crystal. This escape peak is displaced from the photopeak by this discrete amount of energy. Using a similar nomenclature, the so-called Compton Continuum might well be called a Compton Escape Continuum.

The most predominate type of escape peak is due to pair production interaction and its

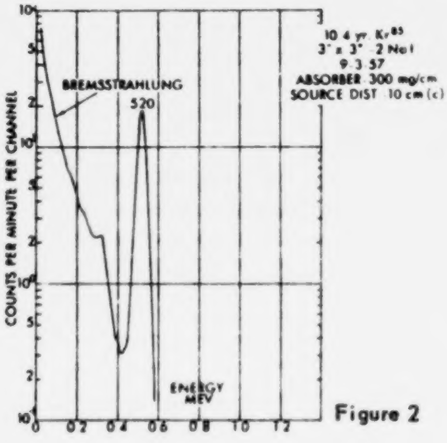


Figure 2

10 4 yr. Kr^{85}
3" x 3" -2 Na I
9-3-57
ABSORBER 300 mg/cm
SOURCE DIST 10 cm (c)

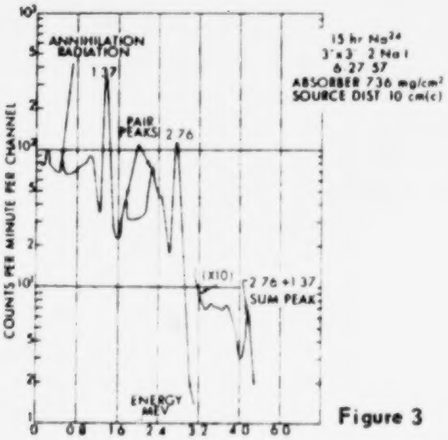


Figure 3

15 hr Na^{24}
3"x3" 2 Na I
6 27 57
ABSORBER 736 mg/cm²
SOURCE DIST 10 cm(c)

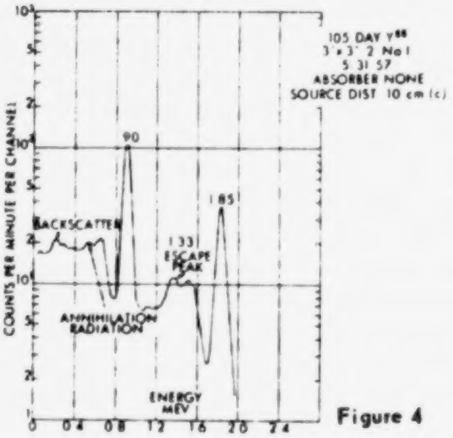


Figure 4

105 DAY Y^{88}
3"x3" 2 Na I
5 31 57
ABSORBER NONE
SOURCE DIST 10 cm (c)

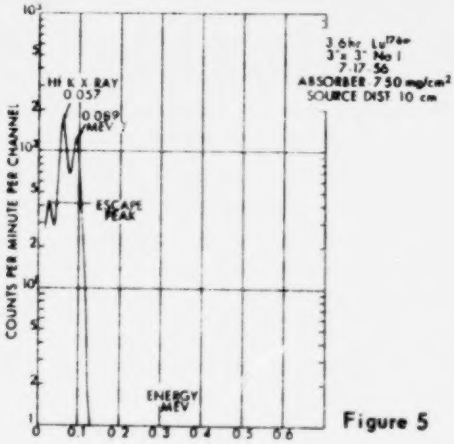


Figure 5

3 6 hr Lu^{176m}
3"x 3" Na I
7-17-56
ABSORBER 7 50 mg/cm²
SOURCE DIST 10 cm

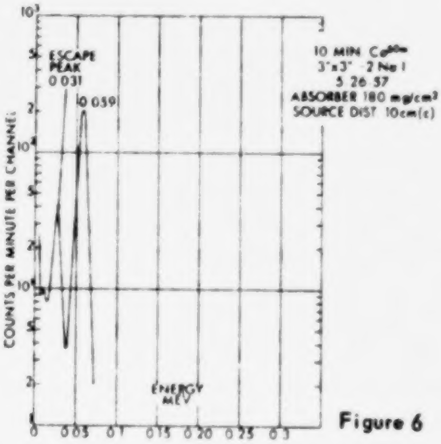


Figure 6

10 MIN Co^{60m}
3"x3" -2 Na I
5 26 57
ABSORBER 180 mg/cm²
SOURCE DIST 10cm(c)

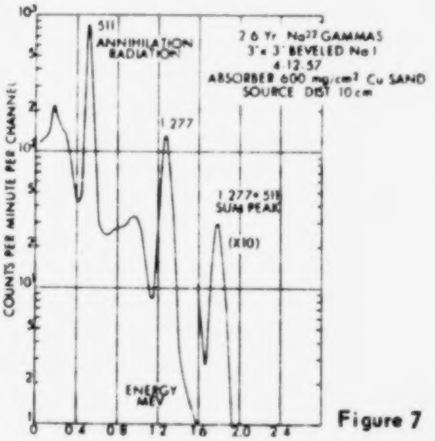


Figure 7

2 6 Yr Na^{22} GAMMAS
3"x 3" BEVELED Na I
4 12 57
ABSORBER 600 mg/cm² Cu SAND
SOURCE DIST 10 cm

subsequent annihilation radiation. When the pair production position interacts with an electron they both disappear and two 0.51 Mev photons are given off at 180^{o} from each other. If this annihilation takes place near the surface of the crystal, one of the two photons may escape the crystal while the other is absorbed. By repetitions of this occurrence, a second peak occurs in the spectrum which is at an energy 0.51 Mev less than the photopeak. Of course, this type of escape peak will only be seen for incident gammas of over 1.02 Mev as pair production will not occur below this energy. There is also the possibility that both annihilation photons will escape, causing a second escape peak to occur at 1.02 Mev less than the photopeak energy. Some examples of this type of escape peak are shown in Figures 3 and 4.

A second type of escape peak is due to the fact that the photoelectric absorption in sodium iodide scintillation crystals occurs mainly in the iodine K-shells. This requires a minimum of 33.2 Kev energy in the incident radiation in order to eject a K-shell electron from the iodine atom. The L-shell electrons in the iodine atom exist at an energy of about 5.2 Kev. These L-shell electrons will drop into 85 percent of the K-shell vacancies with the emission of a characteristic x-ray with energy equal to the difference in energy levels, or 33.2-5.2 = 28 Kev. If this x-ray is emitted near the boundaries of the detector, there is an appreciable probability of its escape. This results in another peak in the spectrum 28 Kev less than the photopeak and is referred to as the iodine x-ray escape peak. The energy of the escape peak may be too low to appear on the spectrum, or at higher incident gamma energies (approximately above 200 Kev), it may be incompletely resolved from the photopeak. Further, the probability of generating an iodine x-ray escape peak declines with increasing energy of the incident radiation. For the most part, this is a result of greater penetration of the more energetic radiation into the crystal before the photoelectric interaction takes place, with the resulting smaller probability for escape of characteristic x-rays originating deeper in the crystal. Figures 5 and 6 show illustration of iodine x-ray escape peaks.

V. SUM PEAKS

Sum peaks is the general terminology for gamma ray spectrum peaks that are the result of more than one single photon interacting with the crystal within a sufficiently short period of time, so that the combined light emissions of the crystal are seen as one single light pulse by the photomultiplier tube.

One common type of sum peak is associated with positron annihilation radiation when a positron-emitting source is counted in a well crystal. In this case, the two back-to-back 0.51 Mev gamma photons may both interact, resulting in a spectrum peak at 1.02 Mev.

Some of the 0.51 Mev photons may escape, resulting in both a 0.51 and a 1.02 Mev peak from positron emitters. With a flat crystal there is little probability that a 1.02 Mev peak will occur, and the 0.51 Mev peak will be the only indication of a positron emitter.

A second type of sum peak results from gammas that are emitted in cascade. This is the case, as seen on decay schemes, where one energy transition by gamma emission is directly followed by another gamma transition. These two emissions are so close together that the detector system will see them as one, and will emit a pulse that is proportional in height to the sum of the gammas in cascade. Figure 3 shows an example of this type of sum peak.

The third type of sum peak is due to simultaneous interaction of different gammas from the sample. The probability of such simultaneous interactions is increased both by higher activity of the gamma emitting nuclides and by increasing the crystal size. Figure 7 shows a sum peak resulting from a 1.277 Mev gamma and positron annihilation.

VI. NON-GAMMA COMPONENTS OF SPECTRA

Other types of radiation than gamma rays may cause additional components to be added to a gamma spectrum. The positron emitter is an example of this type that has already been mentioned.

High energy beta particles may make an appreciable contribution to a gamma spectrum. The major type of contribution is the continuous x-ray spectrum called "bremsstrahlung", or "braking radiation", which is emitted when an electron passes through the electric (coulomb) field of a nucleus. This is shown in Figure 2. A second contribution from beta particles is the characteristic x-rays resulting from ionization of atoms and the subsequent shell transition of electrons. Spectrums with characteristic x-rays of daughter products are shown in Figures 1 and 5.

VII. SUMMARY

Each of the elements of a gamma spectrum have been discussed as individual cases. In an actual spectrum where more than one gamma emitter is present, the situation can be quite complex. Some of the common complexities are the superimposing of a photopeak on a Compton Continuum, and the overlapping of similar energy photopeaks. Even in the case of a single nuclide, the situation may be quite complicated. It is not unusual(3) to resolve four peaks in a spectrum from a single gamma transition; i.e., the full energy photopeak of the nuclear transition, the K-shell characteristic x-radiation of the source due to the internal conversion process, and the iodine escape peak associated with each. All of the factors mentioned must be given careful consideration when analyzing a gamma ray spectrum.

The spectrum characteristics that have been discussed in this lecture, will determine the shape of the spectrum which is independent of sample activity. A change in activity will make a proportional change for all energies in the spectrum with the shape remaining constant.

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GPO 883-384

The Detector System

I. INTRODUCTION

The purpose of the scintillation detector is to convert incoming gamma radiation into electronic pulses which may be counted or analyzed by means of a gamma spectrometer. The basic parts of the detector, as shown in Figure 1, are the scintillation crystal, the reflector, the crystal container, the window and the photomultiplier tube which contains the photocathode. In order to discuss the process of detection, the operation of the detector can be divided into six consecutive events.(1,2)

A. Absorption of gamma radiation in the scintillation crystal

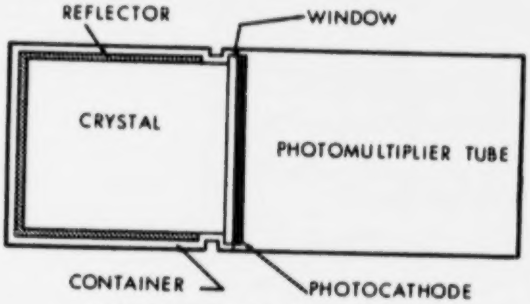
B. Excitation of crystal

C. Emission of light by the excited crystal

D. Transit of this light to the photocathode

E. Emission of electrons by the illuminated photocathode

F. Multiplication of these electrons in the photomultiplier tube



SCHEMATIC DIAGRAM OF A SCINTILLATION DETECTOR
Figure 1

II. SCINTILLATION PROCESS

The process by which the detector crystal converts incoming gamma radiation into light consists of two steps. First, the gamma radiation is converted into energetic electrons by the well known processes of photoelectric emission, compton scattering, and pair production.(7) Second, these electrons cause the crystal to emit light.

A. Luminescence

The absorption of energy by the crystal and subsequent emission of light in the visible or near-visible region, is referred to by the general term luminescence.(1) If the emission of light is within 10^{-8} seconds after the crystal is excited, it is called fluorescence. If it takes longer, it is called phosphorescence.

B. Model of Luminescence

The process of luminescence is not yet completely understood but an outline of the generally accepted theory for the case of activated inorganic crystals can be given.(1, 3, 4, 5)

1. FORMATION OF EXCITONS

The energetic electrons resulting from the interaction of gamma radiation with the crystal, lose their energy by knocking electrons from the outer or valence shells of the crystal atoms, thus ionizing them. Sometimes the valence electrons receive less energy than is required for ionization and may then form an exciton. An exciton is a close coupling of the electron and the hole it left behind in the electron structure of the crystal. The electron is attracted to this hole because the hole is the lowest energy state of the electron and all matter tends toward lower energy. The exciton is somewhat analogous to the hydrogen atom where the electron is attracted to the nucleus but is held in place by its orbital energy. Two important properties of excitons are that they contain energy and are free to migrate through the crystal.

2. ACTIVATORS

The activator atoms are impurities which are intentionally added to the pure crystal to increase its luminescent properties. In Figure 2 it is seen that for the common case of sodium iodide, the addition of small amounts of thallium greatly increases the light output of the crystal.(2)

3. DIFFUSION

The excitons diffuse through the crystal until they are captured by the activator atoms or other energy traps in the crystal. The excitons may lose their energy to other atoms besides the activators or to thermal vibration.

4. EXCITATION

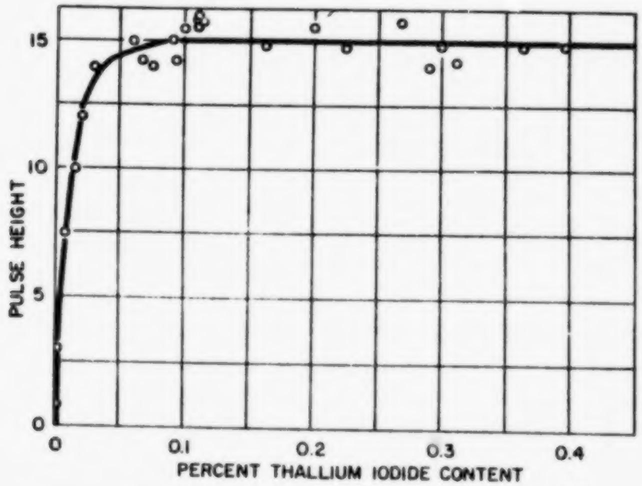
If the activator atom captures the exciton then it is raised to an excited energy state by the energy of the exciton.

5. RADIATION

The excited activator atom may then radiate its energy in the form of light.

III. THE SCINTILLATION CRYSTAL

A great variety of materials will emit scintillations when energy is imparted to them. These materials are usually in the form of inorganic crystals and organic liquids, plastics and crystals. Of these, NaI(Tl), an inorganic crystal, is most commonly used for low-level environmental gamma spectroscopy. Table 1 lists the properties of several different scintillators for comparison with NaI(Tl).



Effect of thallium content on pulse height of the Cs^{137} photoelectric peak recorded by 1" dia. × 1/2" thick thallium activated sodium iodide crystals. (J.A. Harshaw, E.C. Stewart, J.O. Hay; AEC Report NYO 1577; 1952.)

Figure 2

Table 1. PROPERTIES OF SCINTILLATORS

Type	Scintillator	Density g/cm^3	Wavelength of Maximum Emission Å	Decay Constant μ Sec.	Relative Pulse Height	Index of Refraction	Effective Atomic Number
Inorganic Crystal	NaI(Tl)	3.67	4100	.25	210	1.75	50
	CsI(Tl)	4.51	--	1.1	55	1.79	54
	KI(Tl)	3.13	4100	1	50	1.68	49
	LiI(Eu)	4.06	4400	1.4	74	1.95	52
Organic Crystal	Anthracene	1.25	4400	.032	100	1.59	5.8
	Trans-Stilbene	1.16	4100	.006	60	1.62	5.7
Plastic	Terphenyl in Polystyrene	1.06	4000	.005	39	1.59	(low)
Liquid	P-Terphenyl in Xylene	.87	3700	.007	45	1.50	(low)

(5) (Ref. 6, 19)

A. Density and Atomic Number

Because of the great penetrating power of gamma rays, a material of high density and atomic number is required to absorb them efficiently.(3) Inorganic crystals, particularly NaI(Tl) with a density of 3.67 g/cm^3 and an effective atomic number of 50, are superior in this respect to other scintillators. NaI(Tl) can therefore be used in smaller sizes with relatively high detection efficiency.

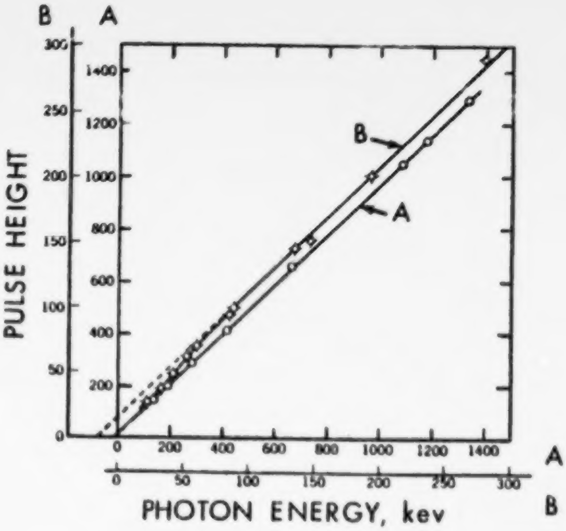
B. Light Yield

The inorganic crystals are superior for converting absorbed gamma radiation into light. This is an important property since the scintillation detection process is statistical in nature which means that the more events that occur the less is the standard deviation of the measurement.

C. Pulse Height

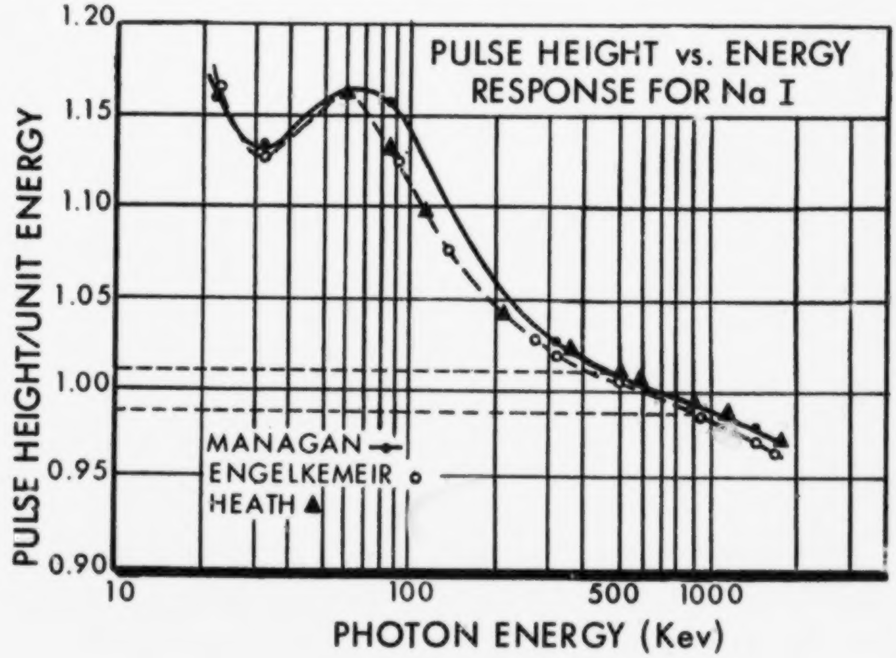
Pulse height is a measure of the light energy produced by the gamma ray in the crystal. After multiplication in the photomultiplier tube this light is represented by an electronic pulse whose height is proportional to the amount of light. A large pulse height per unit of energy absorbed is desirable in a scintillator so that gamma rays of nearly the same energy can be more easily resolved. Merely increasing the gain of the detector will not have the same effect since this also increases the error associated with the measurement of the pulse height.

Figure 3 shows that the response of the NaI(T1) crystal is linear in the energy range above .1 Mev. However, pulse height is never proportional to energy because the straight line does not pass through the origin and the response is particularly nonproportional below .1 Mev. This nonproportional response is accentuated in Figure 4 where the scintillation efficiency (amount of light produced per unit energy) is plotted against the log of energy.(2, 17) A proportional response would be represented by a horizontal line, so considerable nonproportionality is evident and this fact is responsible for a considerable part of the loss of resolution in the detector.(12, 13, 14, 15, 16) As an example, if a 1 Mev gamma ray is absorbed by the photoelectric effect then a certain amount of light is produced as seen in Figure 4. However, if the gamma ray is absorbed by a .5 Mev Compton



PULSE HEIGHT VERSUS ENERGY FOR GAMMA RAYS ON NaI(Tl)(Ref. 3)

Figure 3



NONPROPORTIONALITY OF NaI(Tl)(Ref. 2)

Figure 4

scattering event and .5 Mev photoelectric event, then more light is produced. Thus, gamma rays of the same energy can be totally absorbed in different ways to produce different amounts of light and therefore different pulse heights.

D. Transparency

To be useful, a scintillator must be transparent to the light it emits. The emitted radiation should travel from the point of origin to the exit face or window, essentially unchanged. Practically speaking NaI(T1) crystals are 100% transparent to their own light.

E. Emission Spectrum

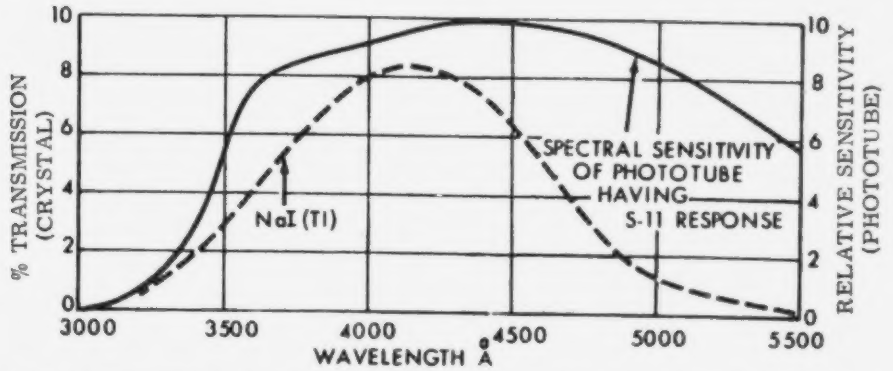
The emission spectrum of a scintillator is the wavelength spectrum of light which it emits in the luminescent process. Figure 5 shows the emission spectrum of NaI(T1) which is centered in the blue portion of the light spectrum and lies within the range of sensitivity of a typical photocathode having an S-11 response. The S-11 response classifies the photocathode as to what wavelength of light will excite it.

F. Physical Stability

Different scintillators have their own particular physical weaknesses. For example, NaI(T1) is very hygroscopic and must be completely protected from moisture or the surface of the crystal will become opaque and the resolution of the detector will be degraded. It is also relatively soft and can be scratched by facial tissue.(2, 23)

G. Decay Time

The light emitted by many scintillators following excitation by gamma rays is found to decay with an exponential dependence.(3) The decay time is defined as the time required after the arrival of a gamma ray for 63% of the light photons to be emitted.(1) This determines how soon another pulse can arrive without overlapping the first and therefore determines the count rate that the crystal can handle without pile-up of pulses. The decay time of NaI(T1) is 0.25 microseconds and from Table 1 it is seen that in general the inorganic crystals are much slower than other scintillators. This is not a great disadvantage in environmental work since faster decay times are not needed. For example, common



Emission Spectra of Anthracene and Thallium Activated Sodium Iodide Crystals (P.R. Bell, R.C. Davis, J.E. Francis, J. Cassidy; ORNL Progress Report 1092; 6/20/51). This emission is covered adequately by a phototube with S-11 response. (Courtesy of Radio Corporation of America)

Figure 5

multi-channel analyzers cannot accept count rates higher than NaI(Tl) can actually deliver.

H. Index of Refraction

Since the light produced in the crystal must pass through a glass window to reach the photocathode, it is important that the difference between the index or refraction of the glass and the crystal be as small as possible in order to minimize the reflection at the crystal-window boundary. Minimizing the reflection will result in higher light collection efficiencies at the photocathode. The index of refraction of NaI(Tl) is 1.75. This is quite high and makes reflection at the boundary an important factor in the design of the detector.

I. Temperature

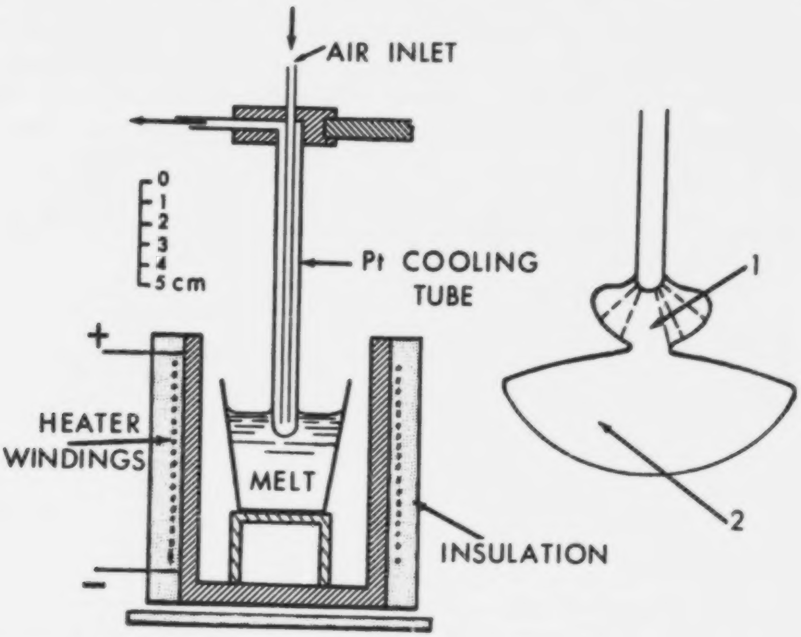
Temperature is observed to affect the pulse height and decay time of scintillators.(3, 8, 11, 24) In general, the pulse height is observed to increase with decreasing temperature for the inorganic crystals and the decay time increases with decreasing temperature. However, the changes observed over the 65-80° F. temperature range are very minimal.

IV. MANUFACTURE

In order to achieve uniform scintillation response, maximum density and optical transparency, it is necessary to prepare NaI(Tl) in the form of a single crystal. The two most commonly used methods of preparing the crystals are the Kyropoulos Method and the Bridgman-Stockbarger Method.[3]

A. Kyropoulos Method

Figure 6 shows a schematic diagram for the production of crystals by the Kyropoulos Method. A platinum tube cooled by the flow of



KYROPOULOS METHOD FOR GROWING SINGLE CRYSTALS (Ref. 3)
Figure 6

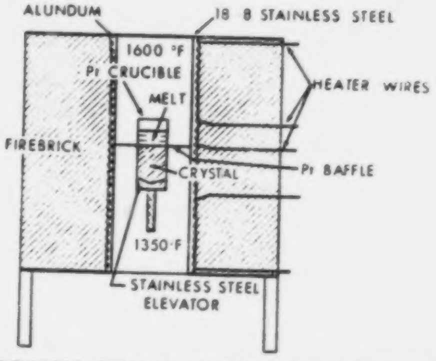
air inside it is lowered into the molten NaI(Tl). When several small crystals have formed on the end of the tube, it is withdrawn until only one crystal remains in contact with the melt. A single large crystal is then grown from the smaller one.

B. Bridgman-Stockbarger Method

Figure 7 shows a schematic diagram of the Bridgman-Stockbarger Method. A two part furnace is used with the top half at a slightly higher temperature than the freezing point of NaI(Tl) and the bottom half at a slightly lower temperature than the freezing point. A sharp temperature gradient is maintained between the two halves by using a platinum baffle. A platinum crucible containing the molten NaI(Tl) is lowered past this temperature gradient to form the crystal. A typical lowering speed is about 1 inch per day.(3)

C. Fabrication

The rough crystal obtained from the manufacturing process is cut to its approximate size by a string saw. Because of the shape of the crucible, this is usually a cylinder. The surface of the crystal is then finished by either grinding or turning on a lathe.



BRIDGMAN-STOCKBARGER METHOD FOR GROWING SINGLE CRYSTALS (Ref. 3)

Figure 7

D. Size Description

The standard crystal is in the form of a right cylinder and is readily available in heights from 1/4" to 6" and diameters from 1/2" to 9".(6) Larger crystals can be specially made but are quite expensive. NaI is easily machined so almost any shape crystal can be made.(2) Common shapes are the well type crystal for high gamma collection efficiencies and the side hole crystal for gamma detection of various flow systems. Examples of several different types of crystals and their containers are given in Figure 8.

V. CANNING

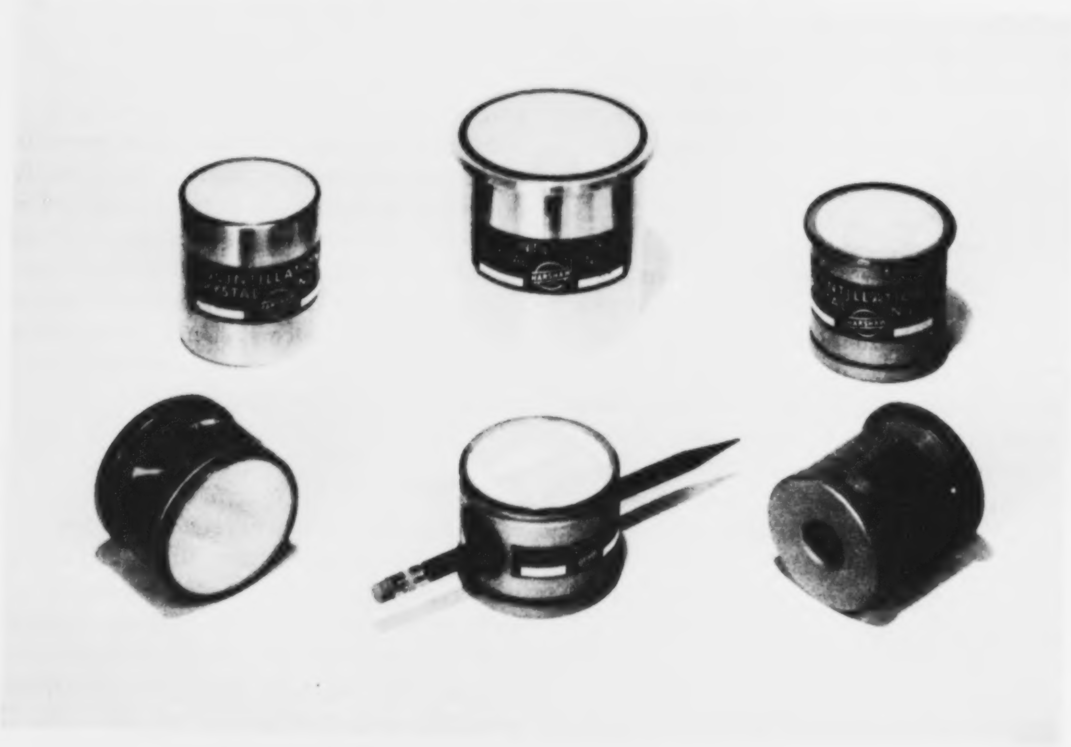
A container for the crystal should provide physical protection, have little interaction with gamma radiation, and efficiently transfer light from the crystal to the photomultiplier tube. Containers are made of aluminum, copper and stainless steel with aluminum being the most common.

A. Physical Protection

Figure 6 shows typical containers for the scintillation crystal. Gamma radiation enters through the sides and top of the can and the light produced leaves through the optical window usually made of glass or quartz. This container will protect the crystal during normal handling and is hermetically sealed to insure that no moisture reaches the crystal.(2)

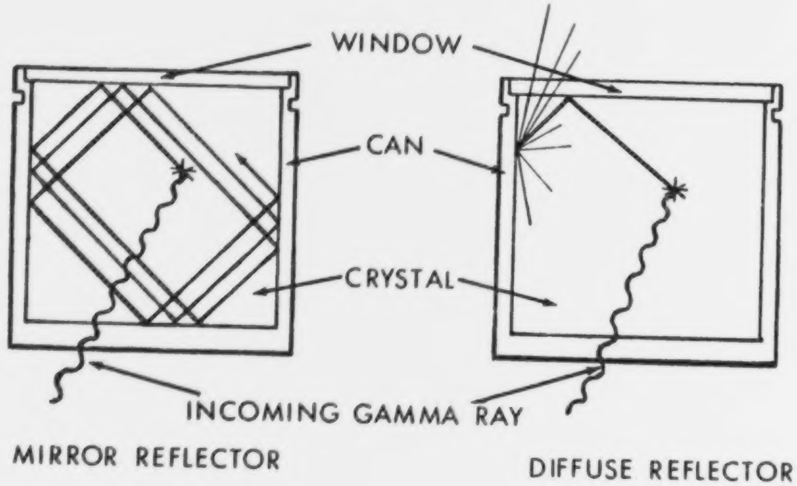
B. Interaction with Radiation

A thin can is necessary so that the incoming gamma rays are affected as little as possible. The standard .032 inch thick aluminum can will absorb less than 1% of the .5 Mev gamma rays. A thin can is also desirable so that Compton backscatter photons are minimized. The can should be of low atomic number to



SCINTILLATION CRYSTALS (Courtesy of Harshaw Chemical Company)

Figure 8



WINDOW
CAN
CRYSTAL
INCOMING GAMMA RAY
MIRROR REFLECTOR
DIFFUSE REFLECTOR

CRYSTAL REFLECTORS

Figure 9

reduce the Bremsstrahlung of incoming betas. The entrance face of the container is sometimes made of beryllium to reduce this effect.(2)

C. Light Transfer

In order to increase the efficiency of the detector, as much light as possible should reach the photocathode from the crystal.

1. REFLECTION

Because of the difference of the index of refraction of NaI(Tl) and common window materials, part of the light incident on the exit face of the container will be reflected back into the crystal.(1) In the standard right cylinder, if the sides of the crystal are mirror reflectors, then light once reflected from the exit face will always be reflected as seen in Figure 9. This, however, will not happen if diffuse reflectors are used for the sides and top of the crystal. Diffuse reflection is achieved by rough grinding the surface of the crystal and then packing a thin layer of highly reflecting MgO or Al_2O_3 on these surfaces before placing in the container.(2)

2. OPTICAL COUPLING

Where light transmission is desired the surfaces in contact are optically coupled using a suitable glue or grease, usually a silicone compound of high viscosity. As an example, the exit face is made by cleaving or polishing one end of the crystal and then optically joining it to the glass or quartz window. By eliminating the air space between the surfaces, the reflection at the exit face is greatly reduced because the light no longer has to pass a boundary where there is a large change in the index of refraction. Problems that arise in optical couplings are air bubbles in the joint which reduce the light transmission and flowing of the coupling material onto reflecting surfaces, thus ruining their reflecting properties.

3. LIGHT PIPE

It is not always possible for the crystal container to be in direct contact with the photomultiplier tube, as is the case for photomultipliers which have curved photocathodes. A light pipe, usually of lucite or quartz is then used to transfer light from the crystal.(1) The best light pipes are made by highly polishing their surfaces.(28)

D. Light Shield

The detector must be shielded from all light entering from the outside because the photocathode is also sensitive to this light. In addition, even a small leak of light from the outside will damage the photomultiplier tube since this light is so much more intense than the light from luminescence. To guard against this, the optical joints are wrapped with black tape to seal out light.(23)

E. Beta Shield

Materials of low atomic number like plastics are often placed between the source and the detectors in order to absorb beta radiation given off by the source.(7) Low atomic number and density are essential to minimize Bremsstrahlung and absorption of gamma radiation respectively. In most cases, however, the aluminum can is sufficient to stop the beta rays from entering the crystal.

VI. PHOTOMULTIPLIER TUBE

Although the photocathode and the electron multiplier are two distinct components, they are always linked together because a vacuum is needed between them and their geometric arrangement is a critical factor in the collection of electrons from the photocathode. Figure 10 and Table 2 show examples and properties respectively of typical photomultiplier tubes.

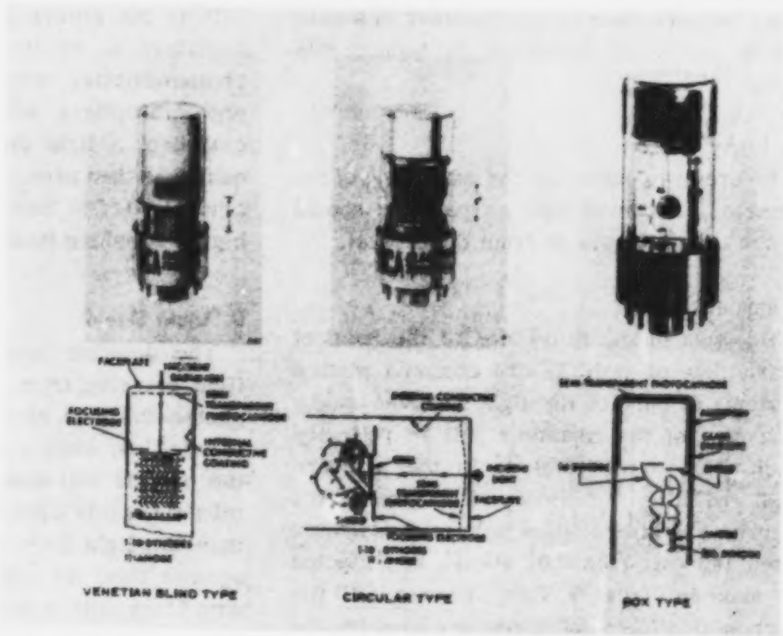

FACEPLATE
FOCUSING ELECTRODE
SEMI-TRANSPARENT PHOTOCATHODE
INTERNAL CONDUCTIVE COATING
1-10 DYNODES
11 ANODE
VENETIAN BLIND TYPE
INTERNAL CONDUCTIVE COATING
FACEPLATE
FOCUSING ELECTRODE
1-10 : DYNODES
11 : ANODE
CIRCULAR TYPE
SEMI-TRANSPARENT PHOTOCATHODE
BOX TYPE

Figure 10 PHOTOMULTIPLIER TUBE
(Courtesy of RCA, Dumont)

A. Photocathode

Photocathodes have the ability to emit electrons when struck by light photons. The most common photocathode is made by evaporating alternate layers of cesium and antimony on the inside surface of the top of the photomultiplier tube.(1, 3) Recently, multialkali photocathodes have been developed which have better temperature characteristics.(27)

1. SPECTRAL SENSITIVITY

Figure 5 shows the range of light to which a typical photocathode having an S-11 response is sensitive. The emission spectra of NaI(Tl) is seen to be within this range. S-11 denotes the response characteristics of the cesium antimony cathode.

2. EFFICIENCY

Of the light photons which reach the photocathode, only about 10% produce electrons.(3) The reason for this low efficiency is illustrated in Figure 11.(27) Much of the incident light is either reflected or transmitted by the photocathode and not all of the absorbed light produces useable electrons. Some light is absorbed producing only heat, some produces electrons with insufficient energy to escape the photocathode and only part of the sufficiently energetic electrons enter the vacuum to be collected by the first dynode.

3. NONUNIFORMITY

The response of the photocathode to light is not uniform over its entire surface.(2) Some parts of the photocathode produce more

Table 2

Characteristics of Representative Photomultipliers

Type	Manufacturer	Photocathode Size (Min. dia. inches)	Spectral Class	Average Sensitivity μa/lumen	Gain	Dia. (in.)	Overall Dimensions (Length, in.)
6342-A	RCA	1.5	S-11	50	2.3×10^{6}*	2-1/4	5-13/16
6655-A	RCA	1.7	S-11	50	2.3×10^{6}*	2-1/4	5-13/16
6810-A	RCA	1.7	S-11	60	6.3×10^{8}**	2-3/8	7-1/2
6199	RCA	1.24	S-11	45	2.8×10^{6}*	1-9/16	4-9/16
6292	Dumont	1.5	S-11	60	2×10^{6}***	2-1/16	5-5/8
6363	Dumont	2.5	S-11	60	2×10^{6}***	3	6-1/8
6364	Dumont	4.2	S-11	60	2×10^{6}***	5-1/4	7-1/8
6291	Dumont	1.25	S-11	60	2×10^{6}***	1-1/2	4-1/4
CL 1012	CBS	1.348	S-11	60	2.25×10^{5}*	1-1/2	4-3/4
CL 1002	CBS	1.745	S-11	60	2.25×10^{5}*	2	5-5/8
CL 1003	CBS	2.703	S-11	60	2.25×10^{5}*	3	6-1/8
CL 1015	CBS	4.250	S-11	70	1.90×10^{5}*	5-1/4	7-3/4
9536B	EMI	1.75	S-11	50	6×10^{5}	2-1/16	5-3/4
9578B	EMI	2.5	S-11	50		3-3/32	6-1/8
9579B	EMI	4.37	S-11	50		5	7-1/2
8054	RCA	2.59	S-11	75	8×10^{4}	3	6-5/16

* 1250 v. supply voltage ** 2300 v. supply voltage *** At 145 volts/stage Ref. 6

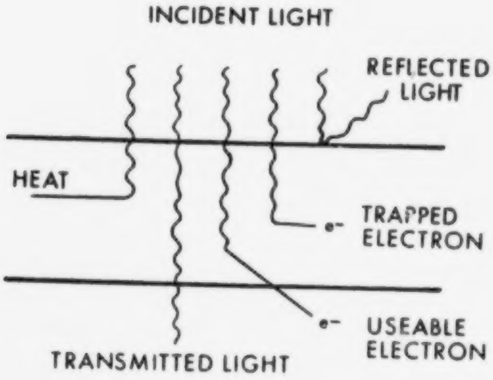
electrons when struck by light than others. Both the light pipe and diffuse reflection from the crystal container, however, help to spread the light uniformly over the photocathode which tends to reduce the effect of non-uniformity.

B. Electron Multiplier

The electron multiplier is designed to multiply the number of electrons given off by the photocathode.(1) Figure 10 shows several different types of electron multiplier structures.(18)

1. COLLECTION OF ELECTRONS

The collection of electrons at the first dynode is the most critical step in the photomultiplier tube. To increase the collection efficiency, grids are placed in the tube to help focus the electrons on the first dynode



INTERACTION OF LIGHT WITH
THE PHOTOCATHODE
Figure 11

and the voltage step to the first dynode is frequently higher than succeeding ones.(1)

2. DYNODES

The electrons from the photocathode are accelerated from stage to stage by placing each successive dynode at a potential of 100 to 150 volts higher than the previous one. The Cs_3Sb or AgMg coatings of the dynodes produce a secondary emission of electrons in excess of those incident. Each dynode multiplies the number of electrons by about four, so that with the usual 10 stages the overall multiplication is about one million.

3. TRANSIT TIME

The transit time is the time it takes for an electron emitted by the photocathode to reach the anode of the photomultiplier tube.(3, 21) The difference between transit times of electrons in the same pulse should be as small as possible, especially for photomultiplier tubes used in coincidence work. In this respect, the circular array of dynodes shown in Figure 10 is best because it provides a strong electric field to draw the secondary electrons from the preceding dynode.(1, 20, 21) Proper focusing of electrons on the first dynode is also important. Common spreads in transit times are about 10-20 millimicroseconds.(3)

C. Temperature

Temperature affects the photomultiplier tube primarily through the photocathode.(3, 10, 24) As temperature increases noise due to thermoionic emission from the photocathode increases. In tubes containing cesium, the tube is not useful above 100° C. As temperature decreases, the resistance of the semiconducting photocathode increases and electrons are not emitted easily. Cs-Sb, for instance, is not useful at liquid air temperatures but some of the multialkali photocathodes can still be used. Temperature also affects the photomultiplier tube by shifting the spectrum of sensitivity of the photocathode.

D. Magnetic Shield

The collection of electrons at the first dynode is very sensitive to magnetic fields.(1, 2, 3) Even the earth's field is sufficient to cause trouble, but it usually does not require the use of a shield because it is a weak field and the detector is not moved through it. To protect against a magnetic field a shield of mu-metal, a material of high permeability made of iron, cobalt and nickel, may be placed around the photomultiplier tube. As a general rule, the shield should extend about one diameter beyond the area to be protected in order to be effective.(2)

E. Pulse Height Shift

The pulse height from the scintillation detector is observed to change with count rate, the pulse height itself and the past history of the detector.(9) The shift is caused by a change in the secondary emission ratio of the dynode surfaces of the photomultiplier tube.

The shift in pulse height is of the order of several percent for count rates which differ by factors of 10 or 100 and usually disappears immediately when the original count rate is restored. However, if very high count rates like 100,000 counts per second are used, then the effect of the pulse height shift may remain for several days.(2) The amount of pulse height shift is also dependent on the energy of the gamma rays, with low energy gamma's causing less shift than those of higher energy. The pulse height shift will vary with different types of photomultiplier tubes and even among tubes of the same type so that it is necessary to determine the shift for each individual tube.(3)

F. Fatigue

A pulse height shift which is not reversible is called fatigue.(7, 22) It is caused by the cascading electrons which are energetic enough to knock cesium from the surfaces of the dynodes. This decrease in the cesium content decreases the secondary emission ratio which in turn decreases the pulse height.

G. Noise

Noise in the photomultiplier tube is due mainly to emission of electrons from the photocathode between light pulses, called dark current, which is due to thermoionic emission.(7) This problem is greatly reduced by cooling the photomultiplier tube or by having two photomultiplier tubes view the same crystal and rejecting all counts which are not in coincidence.(9) Noise is usually not a problem except in low energy work because the pulses are only a few Kev in energy and can be discriminated against.

VII. SUMMARY

The detection of a gamma ray in the scintillation counter starts with the absorption of the ray by the crystal. This produces energetic electrons which excite the crystal. The excited crystal then emits light and this light travels to the photocathode. The transit of light to the photocathode is aided by the use of diffuse reflectors and optical couplings which prevent light from being lost in the crystal. When the light photons strike the photocathode they cause it to emit electrons. These electrons are then drawn to the dynode system which multiplies them about a million times so that they are easily detected.

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The Pulse Analyzer System

I. INTRODUCTION

One distinguishing factor between gamma scintillation systems and gas-filled systems is the method of dealing with the resulting electrical pulse. In gas-filled systems the pulses produced by the interaction of radiation are "counted", either as individual pulses or as resultant current levels; in gamma scintillation systems the resultant pulses are first "analyzed" and then "counted" according to the individual pulse heights. Since the pulses produced in gamma scintillation systems are proportional to the interacting gamma energy, pulse height analysis affords a means of energy differentiation and subsequent nuclide analysis. Therefore, it is important to understand the operation and workings of the pulse analyzer system in gamma spectrometers.

This discussion will include the system which accepts the pulse from the photomultiplier tube, amplifies and analyzes it, and stores the information so that it will be available for various readout devices. See Figure 1.

II. COMPONENTS OF THE ANALYZER SYSTEM

A. Preamplifier

The electrical pulse produced at the anode of the photomultiplier tube is weak because of the high impedance* for the voltage involved. Frequently, space limitations will dictate that the linear amplifier be placed in a location away from the detector system so that the pulse must be sent through connecting wiring. In order to minimize stray capacitance** associated with the connecting wiring, it is desirable to put the first amplifier stage, known as the preamplifier, as near the detector as possible. The main function of the preamplifier is to convert the weak (high impedance) electrical pulses produced by the detector system into strong (low impedance) pulses capable of being conducted through a cable to the amplifier or scaler.(1) Since the strong pulses are not hindered as much by the stray capacitance present in the connecting cables one of the main functions of the preamplifier is to serve as an impedance-matcher between the photomultiplier tube and main amplifier. Impedance matching allows the transport of the pulse with the least amount of distortion and losses, this being quite important in the overall objective of the analyzer system.

The output current pulse from the photomultiplier tube (input pulse to preamplifier) is usually quite small, and the associated voltage pulse is also small; i.e., 0.01 volts to 0.1 volts. It is desired to make this voltage pulse large for ease in further amplification and analysis. A high signal/noise ratio at the amplifier input tends to give better statistics to the process. In striving for a high

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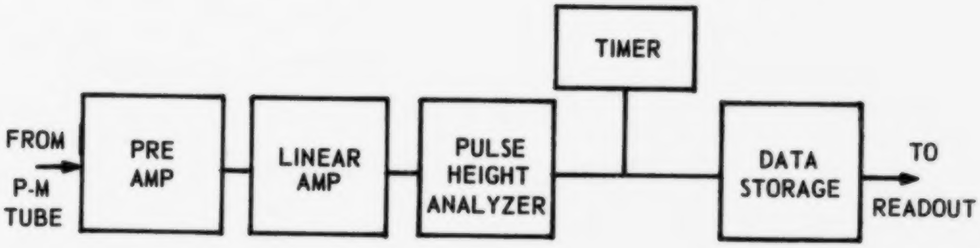
* Impedance is resistance to alternating current flow, being composed of opposition to the flow of current (resistance) and opposition to any change in the flow of current (inductance). It is expressed in ohms.

** Capacitance influences the flow of current by storage, thus altering the flow through a wire.



THE PULSE ANALYZER SYSTEM

Figure 1

signal/noise ratio and matching the impedance, a cathode follower*** preamplifier can be used. The detector pulse is integrated on a very small capacitance, yielding a voltage equal to the pulse charge divided by the capacitance, see equation 1.

$$V = \frac{Q}{C} \qquad \text{(equation 1)}$$

where V = Voltage (volts)

Q = Pulse charge (coulombs)

C = Capacitance (farads)

"The capacitor is made as small as possible in order to make the voltage signal large compared to amplifier noise and to reduce the amount of subsequent amplification required."(2)

The pulses to the cathode-follower preamplifier are several millimicroseconds in width from beginning to end, this width being caused by the Gaussian time distribution. It is desirable to have the integration time of the capacitor sufficient to include the main portion of the Gaussian distribution pulse. Therefore, an integrating time constant of greater than 10 millimicroseconds is large enough for fast scintillators and photomultiplier tubes.(3)

Preamplifier gains (pulse amplification) may vary from less than 1 to 100, the particular gain depending on the designer.

Higher gains in the preamplifier means that lower gains are sufficient in the subsequent amplifier. When the preamplifier has a gain near unity, this being the normal case, it is serving mainly as an impedance-matcher.

B. Linear Amplifier

The overall purpose of the amplifiers is to transform the detector signals into signals suitable for pulse height analysis.(4) This transformation consists primarily of two parts, pulse shaping and amplification. In most instances pulse shaping is done in connection with integrating the detector charge on a capacitor in the cathode-follower preamplifier. Therefore, the main function of the linear amplifier in systems depicted by Figure 1 is to perform amplification. It is desired to have linear rather than nonlinear amplification because of the importance of keeping the relative pulse sizes constant.

The linear amplifier need not be located directly adjacent to the detector system. In fact, space limitations and operational requirements may dictate that it be located a considerable distance away.

Some requirements and desirable characteristics of linear amplifiers are listed as follows:

1. Gain - The gain (pulse amplification) requirements depend on available input

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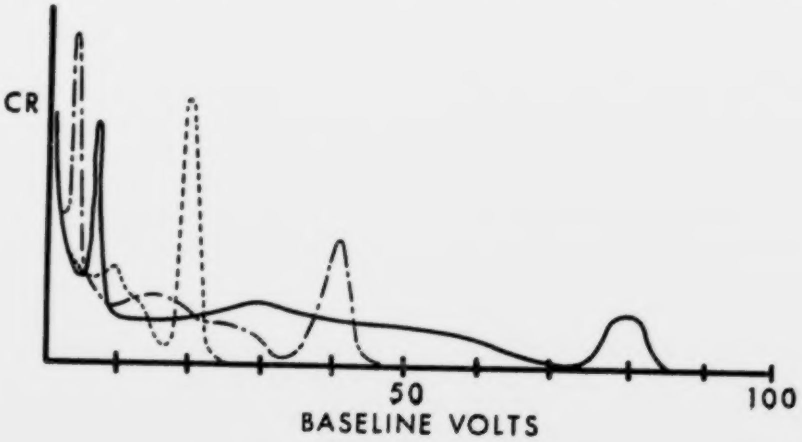
***Cathode follower is one classification of an amplifier circuit, thus indicating that the cathode is common to both the input and output circuits.

signals and the desired output voltages. Since accurate pulse-height analysis work normally requires output voltages up to 100 volts maximum, gain requirements of 10^3 to 10^4 may be required from the photomultiplier tube anode to the analyzer. This gain requirement can be fulfilled jointly by the preamplifier and linear amplifier or by the linear amplifier alone.

Gain control must be provided if the amplifier is to be versatile. Gain control, usually composed of a coarse and fine control capacity, is provided by utilizing helipotentiometers which provide variable resistance and are quite precise. The coarse control usually allows varying the gain binary-wise over a wide range. The fine control should have a variable range overlapping one binary step in the coarse control. The amplifier gain can be used to "calibrate" the instrument. Calibration involves varying the height of pulses representing certain gamma energies to correspond to desired channel numbers. Calibration of the instrument can also be accomplished by varying the high voltage applied to the photomultiplier tube (photomultiplier gain). The effect of varying the height of the pulses is to increase or decrease the channel number (voltage) representing a certain gamma energy on the x-axis of the spectrum, see Figure 2.(5)

A basic step in meaningful gamma spectral analysis is to determine the gamma energy corresponding to a certain channel number.

The stability of the amplifier gain, usually expressed as a percent change as a result of some percent change in line voltage, is very important in gamma scintillation spectrometers. Since proportionality between pulse height and energy dissipated in the crystal is desirable, it is important to keep this constant. Also, in counting, a shift in gain may cause erroneous counts per channel (energy) to be recorded. Therefore, "a change of a small fraction of percent in the output voltage for a 1 percent change in line voltage is not an uncommon requirement. The primary methods employed for



Effect on Cs^{137} differential spectrum of changing amplifier or photomultiplier gain by factors of two.

Figure 2

stabilization of the gain are the use of negative feedback* in the amplifier section and the provision for good regulation in the power-supply section".(6) The gain stability should be independent of other variables; such as pulse frequency, pulse size and shape, time, and threshold setting.

2. Linearity - Linearity in relation to the linear amplifier involves the variation of the ratio between the pulse input and pulse output signal. Linearity in connection with the total system will be discussed in later outlines. The ratio of output pulse height to input pulse height should be constant. However, since this is not entirely possible, limitations must be set. Linearity is expressed as percent of rated output, with a maximum of 0.5 percent often required.(6) This means that the output signal must not deviate from theoretical constant ratio output signal value by more than 0.5 percent. For example, if 50 volts is the theoretical output signal value, the actual value must not be less than 49.75 volts nor more than 50.25 volts. Negative feedback helps in improving linearity.

3. Noise - Noise in a gamma spectrometer is any signal not arising from a nuclear interaction in the detector.(6) Noise can be caused by hum, microphonics, resistor noise, any defective components of the electronic systems, etc.

Typical root-mean-square noise voltages are around 3 microvolts, with rare peaks of maybe 10 microvolts or higher. It is desirable to keep the noise level as low as possible so as to minimize interferences in amplification and analysis.

4. Polarity - The input pulse signal to the amplifier can have either positive or negative polarity, depending on the high voltage supply. Negative polarity is more common for the input pulse to the amplifier, whereas positive polarity is common for the output pulse.

5. Overload tolerance - The amplifier should be capable of recovering from overload pulses, whether they be of peak amplitude, or high frequency.(7)

6. Pulse shape - Linear amplifiers may be categorized according to the output pulse shape, this being defined in terms of five characteristics:

a. Delay time - delay time (T_D) is the time required for the pulse to reach one-half its maximum value. See Figure 3.

b. Rise time - rise time (T_R) is the time for the pulse to rise from 0.1 to 0.9 of its maximum value, this corresponds to the reciprocal of the slope at one half the maximum pulse height and is usually on the order of microseconds.

c. Decay time - decay time (T_{De}), sometimes known as the fall time, is the time for the pulse to fall from its maximum to 0.1 of the maximum.

d. Pulse width - pulse width is a term used to denote the time from rise to fall of the pulse.

e. Pulse height - this is usually considered as the peak height of the pulse expressed in volts.

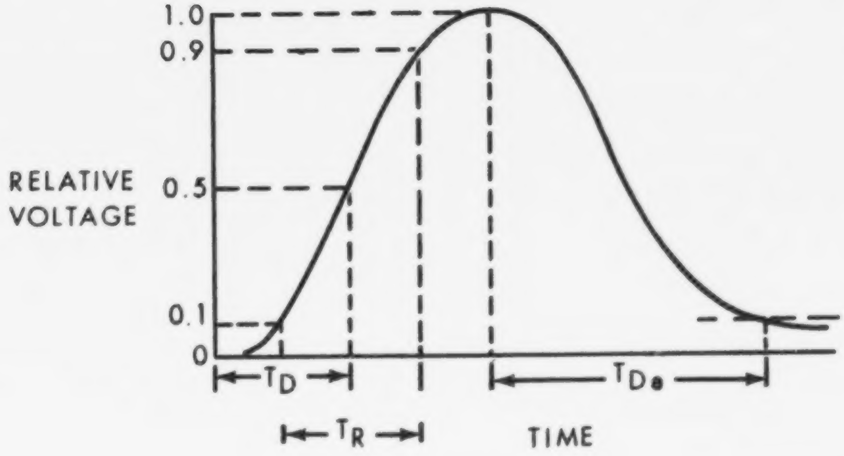
Often only one or two pulse characteristics are listed in the specifications of a linear amplifier.

C. Pulse Height Analyzer

After the pulse leaves the linear amplifier, it enters the analyzer to be sorted according

---

* In negative feedback part of the output signal is "fedback" so as to oppose and decrease the effectiveness of the input signal. As the amount of negative feedback is increased the gain of the amplifier becomes more a function of the feedback network and less dependent on the amplifier and voltage applied.



PULSE CHARACTERISTICS

Figure 3

to the energy it represents. Most pulse height analyzers are voltage sensitive. However, some transistorized analyzers are being built which are current sensitive. Most analyzers measure pulses in the 0 to 100 volt range and with pulse lengths of one to several microseconds.

A common pulse length is approximately 1 μ second, with some pulses having durations as short as 0.5 μ seconds and separations between the leading pulse edges occasionally less than 1 μ second.

1. METHODS OF PULSE HEIGHT ANALYSIS

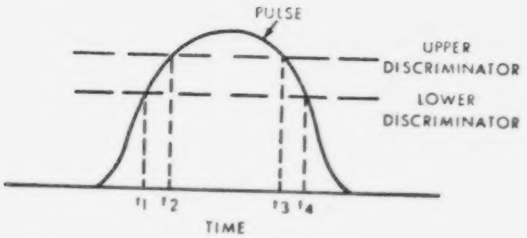
Pulses have been and are being analyzed by many systems, some of which are peculiar to certain instruments and others to many instruments. A mechanical analyzer has been investigated and found inadequate for routine environmental sample analysis.(8) Normally, analyzers can be classified into one of two categories: voltage discriminators or computers.

a. Voltage discriminators

The principle of voltage discrimination is that an output signal is induced only if a pulse height falls in a certain preset voltage range. This principle affords a means of pulse height analysis used extensively in present day "single channel" analyzers. Earlier "multichannel" analyzer designs used this principle; however, present day models are of the computer type.

In the single-channel unit only a narrow band of pulse heights in the x- or gamma spectrum is viewed at any one time, this being accomplished by means of an electronic slit (window width) whose position may be either manually set or controlled by a motor-driven potentiometer. In most single channel analyzers, the output signal from the lower discriminator (base of slit) will be suppressed if a signal arises from the upper discriminator (top of slit) denoting its crossing. The information from the upper and lower discriminator must be stored until after the termination of the input pulse. (See Figure 4)

The pulse is not registered on the lower and upper discriminator at the same time; therefore, this could cause a false output signal from the lower discriminator unless the information arising from this pulse is stored until after it leaves.



VOLTAGE DISCRIMINATOR

Figure 4

Circuits that have an upper level discriminator output signal which can suppress a lower level discriminator signal are called anticoincidence circuits or inhibiting circuits. Some single channel systems register all the pulses (counts) passing the lower and upper discriminators on separate scalers; therefore, lower discriminator scaler counts minus upper discriminator scaler counts equals the counts in the slit (window).

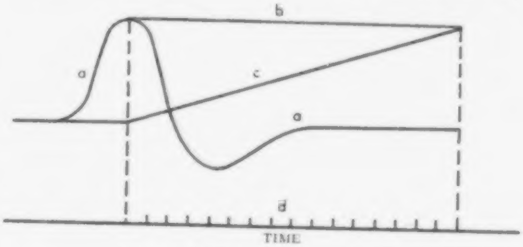
Problems arise with voltage discriminator circuits in relation to setting the windows so that there will be no gap between the adjacent windows (causing loss of counts) and no overlap between the adjacent windows (causing double counting). The instability of channel width (window width) is usually a big limitation on the accuracy of results. The smaller the window width the greater is the error introduced by voltage drift; e.g., a 10 millivolt drift in a 1 volt window would have a greater percentage effect than the same 10 millivolt drift in a 10 volt window. Expander-amplifier circuits have been used to alleviate this problem.(9)

There are two methods of obtaining data from a single channel analyzer; counting in the differential mode and counting in the integral mode. Differential counting involves counting just those pulses which occur within a "window" (between a lower and upper discriminator); whereas integral counting involves counting all pulses above a certain level (lower discriminator).

b. Computer type (analog-to-digital conversion)

Analog-to-digital conversion was introduced in 1949 and has subsequently developed greatly. Advantages offered by this type are many, and include: savings in equipment costs; fewer equipment failures; shorter analysis times, and the ability to achieve completely automatic data readout, both in precision linear graphical and in printed decimal forms. Computer-type pulse height analyzers are used extensively in present day multichannel analyzers.

In analog-to-digital conversion a number is generated in response to each input pulse. This number is assigned in relation to some base number (100 for example) and is called the channel address. These numbers are directed to a computer which causes a count to be stored in the appropriate address in the memory section of the spectrometer. The channel address can be obtained in several ways, including the conversion of the pulse amplitude to an associated time interval and the measurement of that time interval. Time interval measurements are desirable because they can be made with very high precision. There are two essentially equivalent techniques for converting each incoming pulse into a proportional time interval. In Figure 5 "--- an input signal 'a' is converted to a flat-topped signal of the same amplitude 'b' by a pulse-stretching circuit. When the signal has achieved peak amplitude, a linear sweep



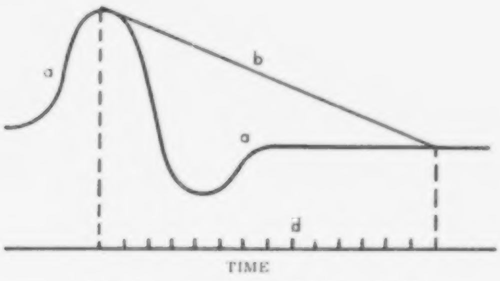
CONSTANT PULSE TIME CONVERTOR

Figure 5

'c' is initiated and, at the same time, a clock oscillator is turned on, generating time marks 'd'. The stretched signal 'b' and the linear sweep 'c' are connected to a comparison circuit which turns off the clock oscillator and resets the stretcher to zero when the sweep attains the amplitude of the stretched signal".(4) The number of time intervals, 'd', corresponds to the channel number of the count to be sent to the computer for appropriate storage. The other major method of amplitude-to-time conversion is shown in Figure 6.

In this method a capacitor is charged to the peak value of the input signal 'a' by a circuit quite similar to a pulse stretcher. However, instead of leaving the capacitor charged to the signal peak, it is discharged with a constant current so that the capacitor voltage decays linearly to zero. The clock oscillator is turned on when the discharge begins and is turned off when a comparison circuit indicates that the capacitor has discharged to its quiescent value.(4) Again the number of time intervals, 'd', corresponds to the channel number at which a count is to be stored in the memory.

Amplitude-to-time conversion methods exhibit channel width uniformity, good linearity, good stability, and relatively simple technical accomplishment.



DECREASING PULSE TIME CONVERTOR

Figure 6

2. SOME PROBLEMS ASSOCIATED WITH PULSE HEIGHT ANALYSIS

Pulses to the analyzer may arrive in a random manner, thus creating problems. The pulses might fall on each other on occasion, causing the electronic circuit to misbehave and register unwarranted counts. Pulses falling on each other, called "pile-up", occurs when more than one pulse arrives during a time interval of the order of the pulse duration. Superposition of small unwanted pulses on either the rising or falling edge of the desired pulse is a form of pile-up.

Another problem in relation to pulse height analysis arises when the pulses have very large variations in height. The presence of cosmic background may cause pulse heights corresponding to several hundred Mev energy. Therefore, problems arise as the analyzer tries to handle these pulses along with other pulses corresponding to normal environmental energies of up to two Mev.

D. Timer

Most spectrometer systems have timer mechanisms associated with them. Timers usually have the flexibility such that preset variable time intervals for sample analysis can be utilized. Some timers have the capacity of indicating either "clock" time or "live" time. There is a certain period of time while the system is analyzing a pulse (order of microseconds) in which it does not have the capacity to analyze another pulse, this time being termed the "dead" time of the system. The actual "clock" time (normal time) minus the "dead" time equals the "live" time (time actually available for pulse analysis).

E. Data Storage

After the signal has been analyzed and "addressed", this information must be recorded or stored in the system until read-out.

Different means can be utilized for the storage of information. Multichannel voltage discriminator analyzers use a separate scaler unit with each "window width" (channel) to accumulate and store data; whereas most single channel analyzers use a single scaler. An advantage of a scaler data storage system is that information can be entered rapidly, in less than a microsecond in some instances. Some of the disadvantages of scalers as a storage system when the number of channels is large are higher costs, higher probability of failure of some component, and difficulty in associating the scaler memory with a read-out device.

In present multichannel analyzers there are two main types of storage facilities used; periodic and nonperiodic.

1. PERIODIC STORAGE FACILITIES

The information can be introduced only at definite moments in periodic storage facilities. The position of these moments on the time axis is periodic, and "the interval between these moments characterizes the quick operation of the storage unit and is called 'memory period'."(10) Magnetic drum memory units and ultrasonic delay line memory units are representative periodic storage facilities.

Normally in data storage a binary numbering system is utilized. There is a certain location in the memory which represents a given pulse height (channel number) and at each channel, spaces for the binary numbering system are located.

Binary numbers are as follows:

Binary	Number
2^0	1
2^1	2
2^2	4
2^3	8
2^4	16
etc.	

Where there is a "1" present in a binary number location or locations at a particular address, this represents that number of pulses analyzed of that particular address; e.g., a "1" at 2^3(8) and 2^1(2) represents a count of 10. Therefore, "1" and "0" are indicative of a "go-no-go" situation.

a. Magnetic drum memory

Magnetic drums are cylinders coated with ferromagnetic material that will retain information stored in the form of magnetized "spots" in appropriate locations. The magnetic drum rotates continuously and when a certain address is to be stored the appropriate spot is magnetized by a "write" coil. The "write" coil puts the new information on the drum and erases the old when necessary; e.g., when going from 15 to 16 counts in a certain address, binary 2^4 would get a "1" and binaries 2^3, 2^2, 2^1, 2^0 would lose their "1" and get a "0" (lose their magnetization). When information is to be read from a particular address the drum, as the portion with the desired address rotates past a "read" coil, induces a voltage in the "read" coil corresponding to the number of counts.

Magnetic drums have high storage capacities because 30-40 binary codes may be located per 1 cm. of drum pathway, with the pathways (addresses) separated by a distance of several millimeters. Due to this capacity, magnetic drums can be used for many-channeled multichannel analyzers. Another advantage is that the information stored does not disappear and is not distorted following disconnection of supplying voltages.

Some disadvantages of magnetic drum memory systems are movable parts, slow "memory period" (order of milliseconds to tens of milliseconds), and slow average "access time"* (order of tens of milliseconds).

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* Access time is that time required to get any desired address data from storage.

Magnetic tapes, operating on the same principles, are being used for both an input storage and output medium.

b. Ultrasonic delay line memory

The first multichannel analyzer using ultrasonic delay line memory was built in 1951. "In general, any device that is characterized by an appreciable time delay between input and output signals can be used as a memory element. It is merely necessary to connect the output to the input through an amplifying and reshaping circuit which compensates for the attenuation and distortion in the delay element. If a signal is introduced into such a system, it continues to circulate indefinitely through the closed memory loop with a period equal to the characteristic delay interval of the delay element. If the delay interval is long, more than one pulse can be introduced at the input before the first pulse returns. The number of pulses that can be stored depends on the time length of delay and on the allowable pulse-packing density, which is determined by the pulse width that the system can pass without excess attenuation and distortion."(11) Because of attenuation and distortion, however, the information capacity of delay lines is not large.

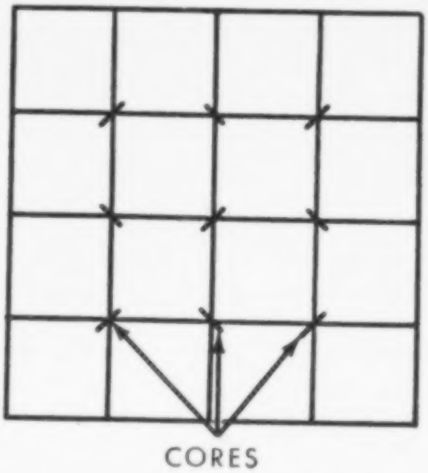
Acoustic delay lines have a larger capacity than electrical delay lines. In an acoustic delay line the pulse is converted to a sound wave which travels back and forth through the medium until it is removed by reconverting to a pulse. Different types of media have been used as acoustic lines, among them being mercury-filled tubes, thin nickel wire, and quartz rods. Different lines in the system correspond to different addresses.

2. NONPERIODIC STORAGE FACILITIES

The information can be introduced at any time into the selected address in nonperiodic storage facilities. Magnetic core storage is the most prominent nonperiodic facility.

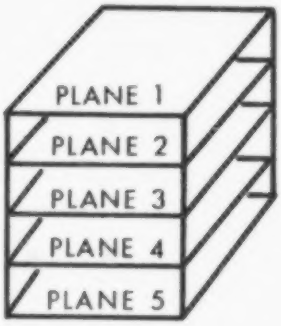
Magnetic core memories consist of doughnut-shaped discs made of a material which can be magnetized by an electrical current. Its magnetic polarity may be in one of two directions, giving it the ability to represent either a "1" or a "0" at a binary location. In magnetic core storage units the cores are arrayed in planes in a sort of herring-bone pattern, see Figures 7 and 8.

The cores in this array have the property that there is a minimal amount of current that must pass through the wires strung



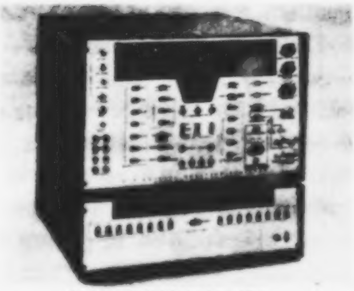
MAGNETIC CORE PLANE

Figure 7



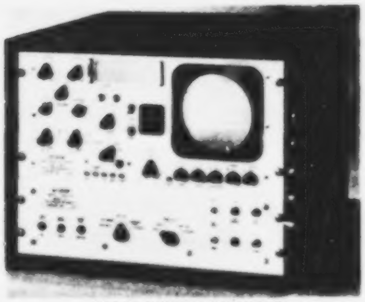
STACK OF MAGNETIC CORE PLANES

Figure 8



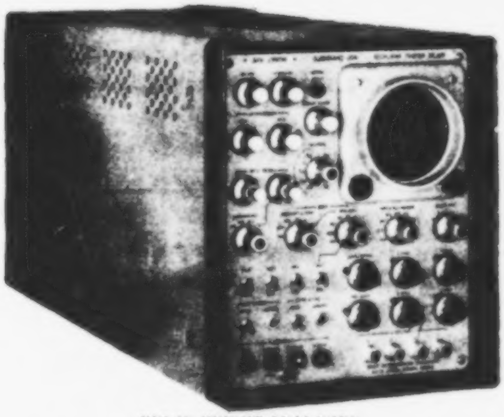
PULSE HEIGHT ANALYZER
"Courtesy of Nuclear Data, Inc."

Figure 9



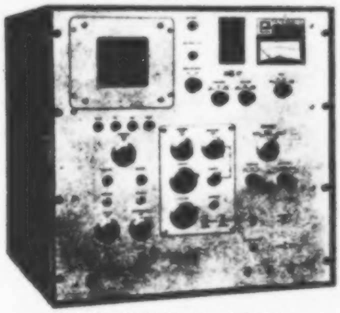
PULSE HEIGHT ANALYZER
"Courtesy of the Victoreen Instrument Company"

Figure 10



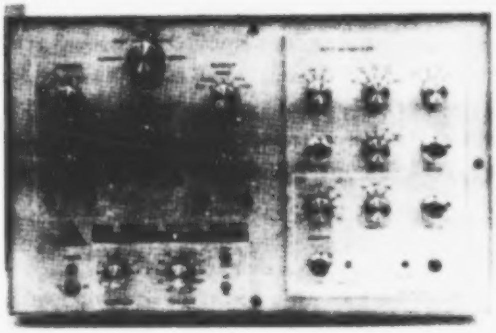
PULSE HEIGHT ANALYZER
"Courtesy of Technical Measurement Corporation"

Figure 11



PULSE HEIGHT ANALYZER
Courtesy of Radiation Instrument Development Laboratory, a Division of Nuclear-Chicago Corporation"

Figure 12



PULSE HEIGHT ANALYZER
"Courtesy of Radiation Counter Laboratories, Inc."

Figure 13

through their centers in order for them to be magnetized, or the magnetic state to be changed. Through any given core in the array there are two "write" wires which serve to set up the magnetic state of the core. When a "1" or "0" is to be stored, one-half of the minimal amount of current is sent along one wire and one-half the minimal current is sent through the appropriate perpendicular so that only at their intersection do the currents in the two wires cause the magnetic state of a core to be changed. Thus, by sending currents in the proper directions along the appropriate row and column of the core array, a "0" or "1" can be stored.

If readout of the counts in a particular core or cores (corresponding to one channel) is desired, a "read" pulse is sent along two or more of the appropriate "write" wires. This read pulse always assumes only one direction for any given core, and attempts to magnetize a core in a specific direction. If the core is already magnetized in this direction, there will be no change in the magnetization of the core. Since there is no change in the magnetic state of the core there will be no voltage induced in the "sense" wire (a wire passing through all the cores). If the core, however, had been magnetized in the opposite direction, the incoming read pulse would change the magnetic state of the core, thus inducing a voltage in the sense wire. This pulse then is read, thus distinguishing between a "0" and "1" in the core by whether or not there is a pulse in the sense wire.

Unfortunately, this type of readout inherently destroys the data contained in the memory. This is undesirable and is overcome as follows:

Each time a read pulse is given in the write wires, there follows a "restoration" pulse through the write wires. In the case where the contents of the core were not destroyed (no pulse is felt in the sense wire), a pulse in the "inhibit" wire prevents or cancels the effect of the restoration pulse.

Magnetic cores have short access times (order of 10 microseconds), and any memory address can be selected at any time. The pulse size from the memory system represents a certain number of counts in a certain channel. The readout device may require that this digit (pulse size) be broken into its binary parts; therefore a serial converter may have to be utilized. This concept of providing an appropriate pulse for a readout device will be discussed elsewhere.

III. SUMMARY

The pulse height analyzer system of a gamma spectrometer is composed of a preamplifier, linear amplifier, pulse height analyzer, timer, and memory section. The operation and inter-relationships of these components can become quite complex. Figures 9, 10, 11, 12, and 13 indicate certain commercially available pulse analyzer systems. Single and multi-channel spectrometers are very useful in the analysis of samples from the various environmental media.

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The Display System

I. INTRODUCTION

The gamma spectral data stored in the memory of the pulse height analyzer is not directly accessible to the operator since the information exists as magnetized and non-magnetized areas in the memory. Therefore, it is necessary to convert this stored information to a form readily interpretable by the operator such as graphic plots or numerical notation, or to a form interpretable by automatic data processing equipment such as magnetic tape or punched paper tape.

Before selecting read-out equipment for any particular application, the application should first be defined and isolated as best as available information will allow. This not only prevents shortcomings in the final use of the equipment, but prevents wasted dollar value; i.e., acquiring equipment whose full potential will never be used. However, additional applications may be realized after initial installation, necessitating additional equipment.

In selecting read-out equipment, the following characteristics should be considered:

1. Input signal form necessary to drive the equipment.

2. Speed of read-out; usually expressed as channels per second.

3. Representation format for numerical output.

4. Accuracy of representation of information stored in memory.

5. Interpretation method to which the data will be subjected.

6. Versatility of controls and applications.

II. DATA REPRESENTATION AND LOGIC SYSTEMS

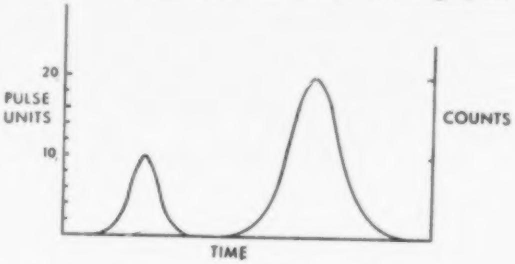
Data from the memory may be represented by either analog or digital output. There is a definite distinction between the two outputs and a read-out device that is compatible with one representation is not normally compatible with the other.

A. Analog

Analog data are presented in a continuous form by means of physical variables such as voltage, resistance, or rotation. If the data are represented by voltage, then a voltage pulse of 10 units will equal 10 counts, a pulse of 20 units will equal 20 counts and so on. Graphically this can be represented as in Figure 1.

In the above case if the total counts in a particular channel were 476, this would be represented by 476 voltage units.

Because the total counts in a channel are directly proportional to the pulse height, an



ANALOG DATA REPRESENTATION

Figure 1

accurate measurement of the pulse height must be obtained. This is done by a voltage measuring device which may or may not be very accurate. This measurement is the main source of error in this type of system.

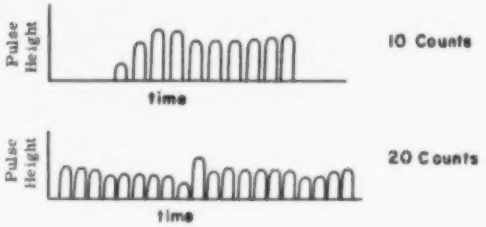
B. Digital

In a digital system, the data are represented in discrete (discontinuous) form usually by coded characters; i.e., numbers, signs, symbols, etc.

In the digital system the number of individual pulses represents the number of counts rather than the counts being a function of the pulse height. If the numbers 10 and 20 are presented in a digital system, 10 and 20 pulses, respectively, would be required. Graphically this can be shown by Figure 2.

As can be seen, the height of the pulse does not determine the number of counts. Although it takes more time to count the pulses, the digital system is more accurate as measurement of the pulse height is not necessary.

In a digital system two logics are used, serial and parallel. A logic is nothing more than a set of instructions that are programmed into the memory, instructing it how to read out the data.

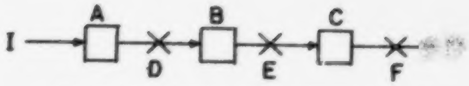


DIGITAL DATA REPRESENTATION

Figure 2

1. SERIAL LOGIC

Serial logic is used when numbers are to be read-out one digit at a time. The following example shows how serial logic is used to retrieve a number from the memory.



INFORMATION RETRIEVAL - SERIAL LOGIC

Figure 3

A - storage location for hundreds digit

B - storage location for tens digit

C - storage location for units digit

D, E - gates (or electronic switches) that open when the preceding storage place is cleared, i.e., turned to zero. This number is then printed out.

F - gate that opens when preceding storage place has been cleared. After opening, the number prints-out, then the retrieval process is advanced to the next channel.

I - Oscillator

P - Print-out

A number, 570, is located in the memory. This is read-out serially by sending in at I an oscillating signal which is a series of individual independent pulses. The oscillating signal will operate on each storage location, one at a time. The operation will be to return the numbers at each storage location to zero if not already so. The number of pulses from the oscillator necessary to return a storage location to zero is equal to the numerical information stored at that location. It will take 5 pulses from the oscillator to turn the A storage location to zero. When this storage location turns to zero a number equal to the number of pulses coming out at "P" will be printed and gate D will open. Then 7 pulses from the oscillator will return the B storage location to zero. When zero is reached, the number is printed and gate E will open. Since the last storage place is already at zero, no pulses are required. Zero is printed and gate F is open which sends the oscillating signal to the next channel where the cycle is repeated until the data at all channel locations are printed out.

2. PARALLEL LOGIC

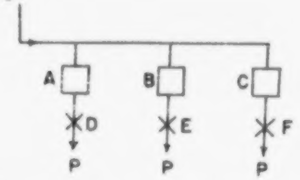
Parallel logic instructs the memory to print all the numbers in a particular channel at once. Again an example is used to illustrate parallel logic.

A number, 570, located in the memory is to be read-out using parallel logic.

The oscillator signals will operate (return to zero) simultaneously on each location, A, B, and C. A separate pulse series for each location is read-out as each location is turned to zero. In the example, only seven pulses would be required to clear all locations.

When all gates are open the total number will be printed and the oscillating signal moves to the next channel where the procedure is repeated until all data are printed out.

Parallel logic is much faster than serial logic. With parellel logic the oscillating signal goes through all storage locations at the same time instead of going through the storage locations one at a time. However, serial logic requires about two-thirds less components than parallel logic, thus the possibility of mechanical failure is less in serial logic.



INFORMATION RETRIEVAL - PARALLEL LOGIC

Figure 4

III. READ-OUT EQUIPMENT

An output format of a given analyzer will drive only those instruments designed to receive it. However, this format may be converted so that the original signal can drive any of a number of various read-out devices. Before discussing converters, it is well to consider the various types of available read-out equipment, classified as to the type of input needed to drive each.

A. Analog

1. CATHODE RAY TUBE (See Figure 5)

The cathode ray tube, better known as an oscilloscope, is an instrument that will give a visual nonpermanent graphical representation of the data. An oscilloscope may or may not be incorporated in the main analyzer unit. A continuous sweep of the memory by an oscillating signal may be incorporated into the accumulation mode of the analysis which permits "live" viewing of the data in the memory.

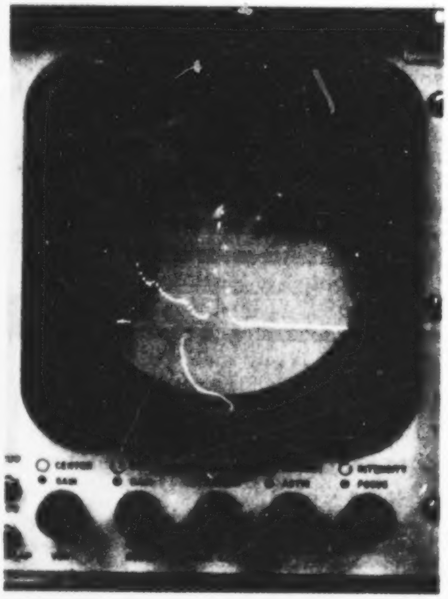


Figure 5

The curve produced has the appearance of "growing" as data are accumulated in the memory.

Advantages of having an oscilloscope on an analyzer are:

a. by watching the graph "grow", an indication of the count rate is given from which a logical counting time may be set

b. gives a quick look at the spectra to determine if any malfunctions have occurred, e.g., channels dropped or shifts

c. it is very useful in the calibration of the gain settings of the instrument.

The major disadvantages of using an oscilloscope for analysis operations are the inaccuracy and non-permanency. Determining the total counts in a particular channel is very difficult making quantitative analysis almost impossible. A camera with rapid developing film has been adapted to take a picture of spectra as they appear on the oscilloscope. This gives a permanent record of the spectra.

The camera is mounted firmly against the oscilloscope and is held by four nuts. The camera lifts in and out of the adapter easily, and the complete unit swings away for unobstructed viewing of the display.

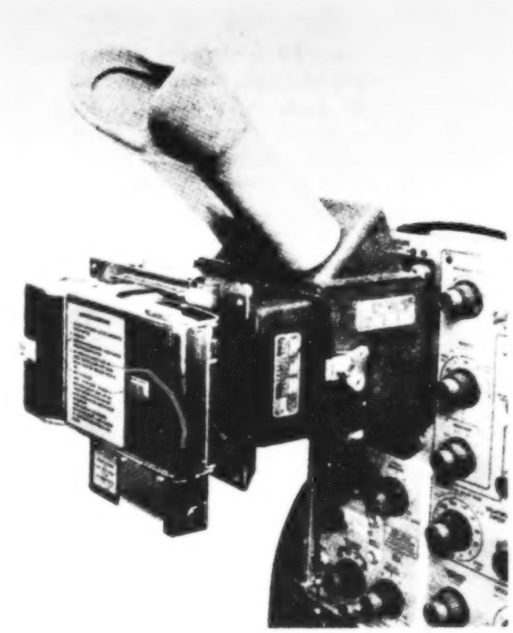
The camera setup has the advantage of being able to store the photographs in a minimum of space since literally hundreds of read-outs can be conveniently sorted and filed in a common office card-index filing cabinet. The photographs can be used for checking standards, for some quantitative work with an accuracy of about 10%, and for qualitative analysis by peak identification.

It is advantageous if an oscilloscope has external controls for:

a. positioning the spectra both horizontally and vertically

b. periodic channel intensifiers for establishing channel position on the horizontal scale

c. expanding the vertical and horizontal distribution of channel points



CAMERA ADAPTION TO THE OSCILLOSCOPE
(Courtesy of the Beattie-Coleman, Inc.)

Figure 6

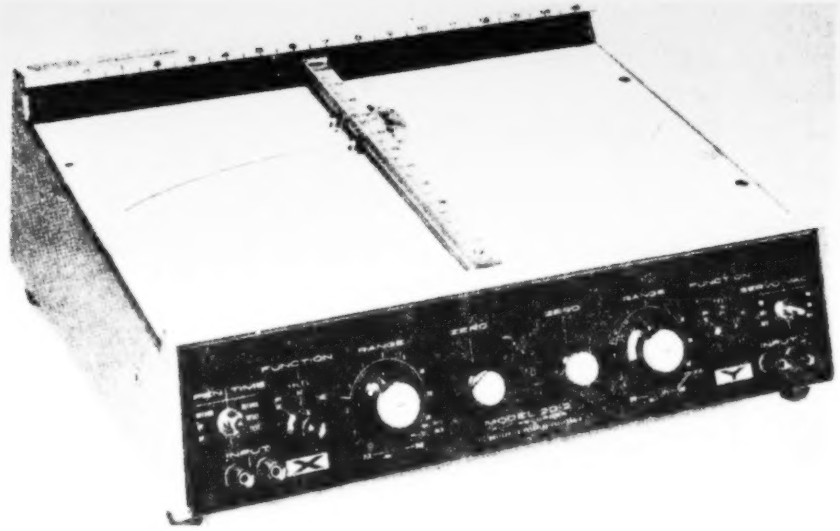
d. increasing or decreasing the vertical scale by steps not greater than a factor of 5

e. focusing and intensifying the spectra points

f. log and linear display.

2. X-Y PLOTTER (See Figure 7)

The basic function of the X-Y plotter is to produce graphic tracings or point plots showing the relationship between two variable functions. Electrical signals representing the functions, channels number and counts in that channel, are applied to the plotter. When the plotter is positioned both as to channel and count, a null detector, which senses that the plotter is positioned, will activate the point plotter to make a point. The carriage then moves to the next channel and count and the plotter again makes a point. When drawing a continuous line spectra, the null detector would not be in operation.



X-Y PLOTTER -

Figure 7 (Courtesy of the F. L. Mesely Co.)

The printout speed of an X-Y plotter is usually a function of how fast the memory reads the information out, although some modifications can be made to increase or decrease the speed. On the average an analyzer will read-out, through the plotter, at a rate of 3-4 channels per second. It is advantageous for a X-Y plotter to have:

a. both fine and course controls for each axis

b. both linear and log plotting capabilities

c. both point and continuous plotting capabilities

d. interchangeable characters for point plotting

e. the capability of using different paper sizes.

The main disadvantage of the X-Y plotter is that the graph produced is not as accurate as hand plotting. This hinders analysis operations but is acceptable if the general shape and approximate values are all that are needed.

3. STRIP CHART RECORDER (See Figure 8)

A strip chart recorder has the basic function of producing inked recordings showing the relationship between a variable function and time. This recorder finds use in single channel gamma analyzers. The variable function normally is count rate or total accumulative counts, while the time base is a representation of the pulse height.

It is advantageous for the strip chart recorder to have the following characteristics:

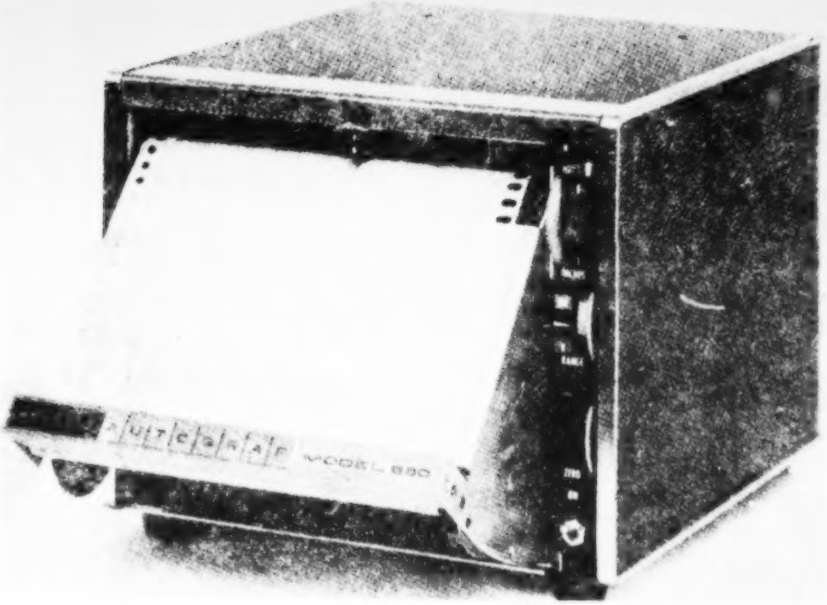
a. The ability to change paper output speeds from about 1 inch per hour to about 8 inches per minute.

b. The ability for locking in different positions for note taking or marking.

c. A drawer for accumulation of paper as it comes out.

d. Rack mounting capability for "building block" assembly.

e. A feed out which is convenient for continuous reading or intermittent tearoff.



STRIP CHART RECORDER - (Courtesy of the F. L. Moseley Co.)

Figure 8

B. Digital - Parallel Logic

1. PARALLEL PRINTER (See Figure 9)

The basic function of a parallel printer is to print the information in numerical form channel by channel, on a paper tape, Figure 10. The output format normally has eleven single digit columns on strip paper about three inches wide. If three columns are used for channel identification, eight remain for the counts in each channel. The format has the advantage that the data can be visually scanned to pick out channels with higher total counts. In some cases this number is reduced to match the maximum capacity of the memory. This will increase the output speed as less wheels have to be activated. The output speed of a printer is on the order of five channels per second but modifications can be made to increase this.

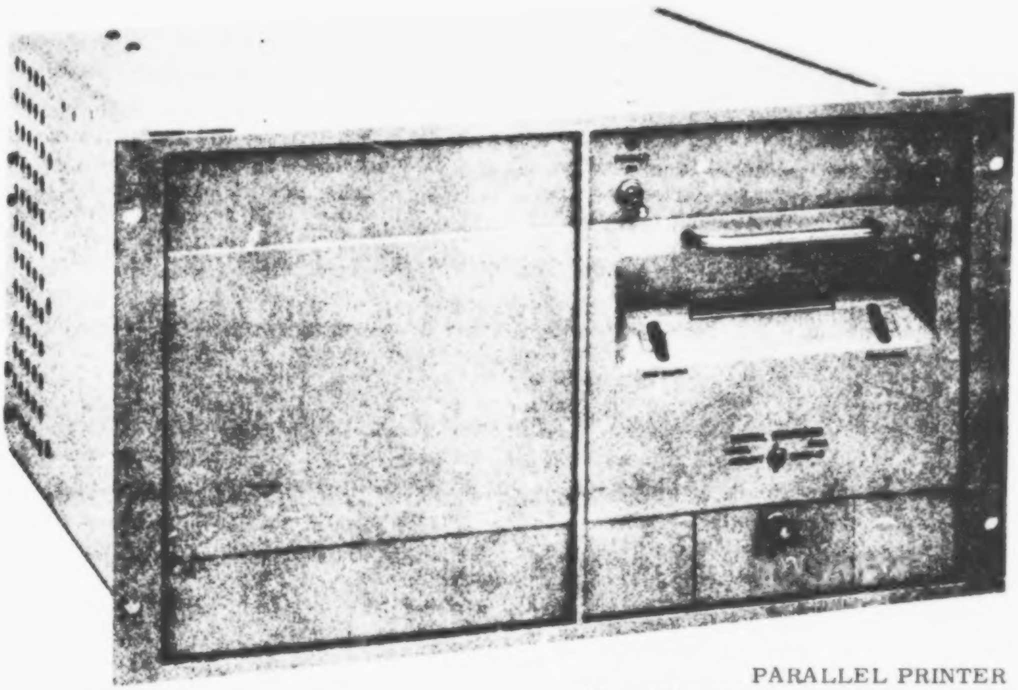
2. SCALERS

A scaler will show the total number of counts that have accumulated in a given channel. The scaler can be set up as a binary system, a decade system, or a combination (binary coded decimal-BCD). The main disadvantage of using a scaler on an analyzer is the manual recording of the data. The output speed of a scaler is limited by how fast the data can be manually recorded.

C. Digital - Serial Logic

1. ELECTRIC TYPEWRITER (see Figure 11)

An electric typewriter is used to type out accumulated information from the analyzer to a standard type written page. Print-out is achieved at a speed of about 12 characters per second, which is about 1.5 channels per second without address identification and 1.1 channels per second with address identification.



PARALLEL PRINTER
(Courtesy of the Hewlett-Packard Company)

Figure 9

```
006        00549
005        00691
004        00474
003        00106
002        00067
001        00000
```

PARALLEL PRINTER TAPE FORMAT

Figure 10

ELECTRIC TYPEWRITER
(Courtesy of International Business Machines Corp.)

Figure 11

Accumulated information is presented in ten-5 digit (channel) groups per line (without channel address identification) or five (channel/data) groups per line (with channel address identification) on the standard 8 1/2 inch X 11 inch paper, Figure 12.

The typewriter has an added chacteristic in that it can be used manually. This provides the capability to type pertinent information on the data sheet, i.e., sample number date, counting time, etc.

2. PUNCH TAPE (see Figures 13 and 14)

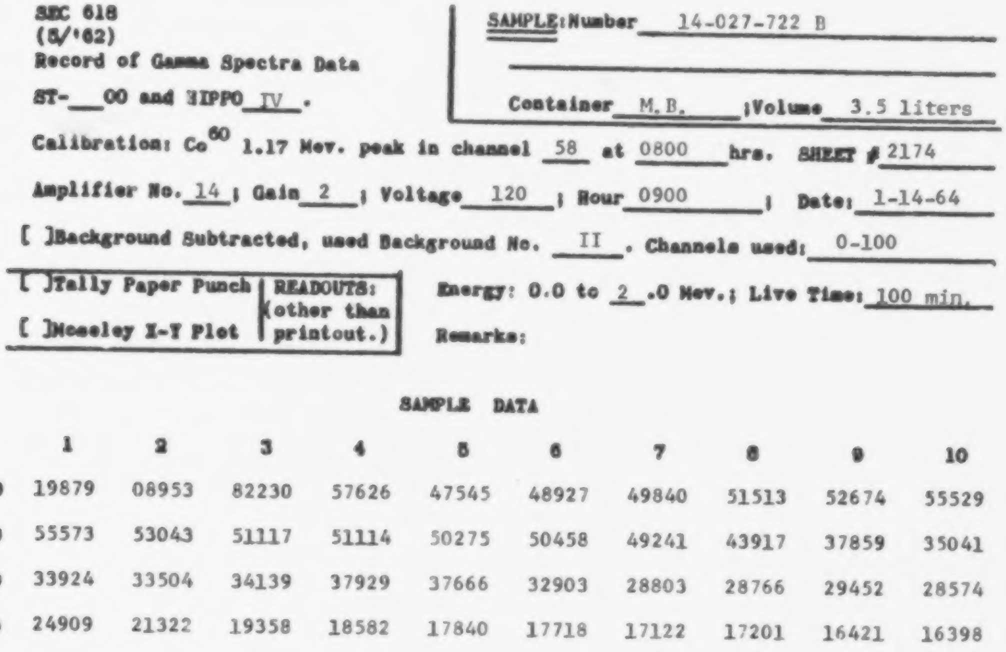
The paper punch tape units are used as both perforator and readers. The perforator unit provides a fast and efficient means of reading-out accumulated information on BCD punched paper tape. Punched paper tape is about 1 inch wide in which holes are punched in a coded system as shown in Figure 15. The basic speed of the perforator is on the order of 60 characters per second which allows approximately 10 channels per second to be printed without channel identification or 6 channels per second with channel identification.

The reader unit can be used to perform four very important basic functions and add appreciably to the versatility of the system.

a. Read information, such as backgrounds, from a previously punched paper tape into the analyzer memory.

b. Read information from a previously punched paper tape into an electric typewriter. The analyzer is free to accumulate new information during this function, since only the serial converter is required to accomplish this step, thus doubly benefiting the user.

c. Read information from a previously punched tape to a paper tape perforator to obtain duplicate tapes.



SEC 618
(5/'62)
Record of Gamma Spectra Data
ST-___00 and SIPPO IV **.**

SAMPLE: Number 14-027-722 B

Container M.B. **; Volume** 3.5 liters

Calibration: Co^{60} **1.17 Mev. peak in channel** 58 **at** 0800 **hrs. SHEET #** 2174

Amplifier No. 14 **; Gain** 2 **; Voltage** 120 **; Hour** 0900 **; Date:** 1-14-64

[ ] Background Subtracted, used Background No. II **. Channels used:** 0-100

[ ] Tally Paper Punch
[ ] Moseley X-Y Plot
READOUTS: (other than printout.)

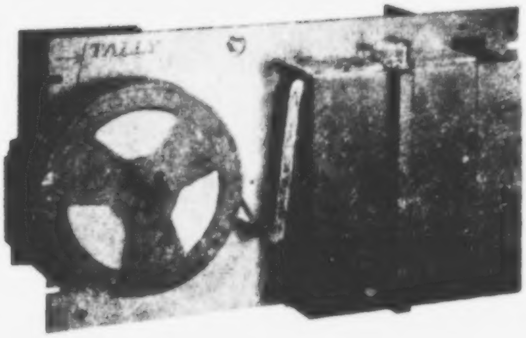
Energy: 0.0 to 2 **.0 Mev.; Live Time:** 100 min.

Remarks:

SAMPLE DATA

	1	2	3	4	5	6	7	8	9	10
00	19879	08953	82230	57626	47545	48927	49840	51513	52674	55529
10	55573	53043	51117	51114	50275	50458	49241	43917	37859	35041
20	33924	33504	34139	37929	37666	32903	28803	28766	29452	28574
30	24909	21322	19358	18582	17840	17718	17122	17201	16421	16398

TYPEWRITER FORMAT
Figure 12



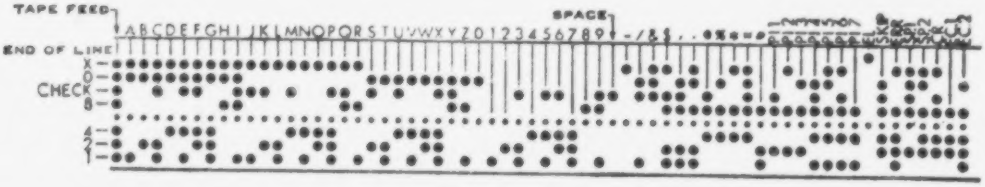
PAPER TAPE PERFORATOR

Figure 13

PAPER TAPE READER

Figure 14

(Courtesy of the Tally Corporation)


TAPE FEED
SPACE
END OF LINE
X
0
CHECK
8
4
2
1
A B C D E F G H I J K L M N O P Q R S T U V W X Y Z 0 1 2 3 4 5 6 7 8 9

PAPER PUNCH TAPE - EIGHT CHANNEL CODE

Figure 15

d. Read information from a previously punched tape simultaneously to a paper tape perforator and electric typewriter, at typewriter speed.

The read-in speed in each case will depend on the speed of the unit into which the information is being used.

A major disadvantage of using paper tape is that the tape is easily torn.

3. MAGNETIC TAPE (see Figure 16)

Magnetic tapes are very similar to paper punch tapes in their use. Instead of punching holes in the tape, the data are recorded on the tape as magnetized spots or bits located along the length of tape, Figure 17.

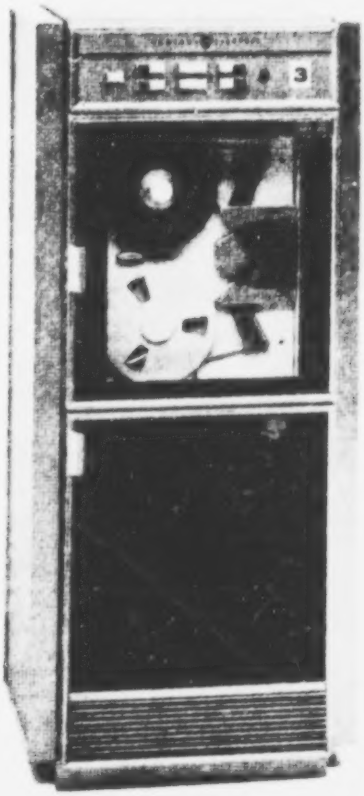
Magnetic tape units are more applicable where large volumes of data are being processed because of the high output speed.

The speed at which the tape will accept information is governed by the output speed of the device relaying the data.

Magnetic tapes have the capacity of storing large volumes of data on a small amount of tape. However, magnetic tape has the disadvantage of being easily torn, losing its magnetism, and difficulty may be experienced in aligning the tape.

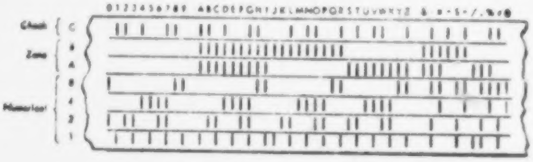
IV. CONVERTERS

Converters have been developed to convert from one system or logic to another. There are analog to digital, digital to analog, serial to parallel, and parellel to serial converters available. Some instruments have a converter as an integral part of the total unit.



MAGNETIC TAPE UNIT
(Illustration reproduced with permission from the General Electric Computer Department)

Figure 16



MAGNETIC TAPE - SEVEN BIT CODE

Figure 17

For instance, if an analyzer is designed with digital read-out using parallel logic, a parallel to serial converter would be necessary to operate an electric typewriter.

V. SUMMARY

Analyzers are setup on two types of output systems, analog and digital. Each system has the ability to drive particular types of print-out devices. Because print-out instruments will only accept a certain type of signal, converters have been developed which allow switching from one system or logic to another.

GPO 884-547

Energy Calibration

I. INTRODUCTION

The data accumulated by a gamma spectrometer is displayed in terms of the total counts per channel vs tne channel number. The value of the baseline, i.e., channel number, has no real significance however, until it can be calibrated proportionally to read in terms of energy, specifically the energy transferred to the crystal by the interaction of the incident gamma rays. Unfortunately, the pulse height received by the analyzer does not always display a proportional, or even linear, relationship to the energy of the incident gamma photon. In addition, a spectrometer system may display instability. In other words the voltage gain (which determines the proportionality factor) may not remain constant. Both nonlinearity and instability must be recognized when calibrating an instrument and their effect minimized where possible.

II. CHANNEL - ENERGY RELATIONSHIP

Before discussing the channel number - gamma energy relationship involved in spectral interpretation, it is necessary to define several terms.

A. Definitions

1. Proportional Response - A proportional response is one that can be represented by the function $y = mx$, which is a straight line passing through the origin. This is shown in Figure 1.

2. Linear Response - A linear response is one that can be represented by the function $y = mx + b$, which is a straight line but does not pass through the origin. This is shown in Figure 1.

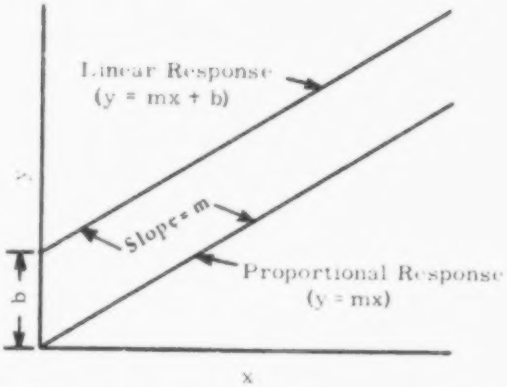


Figure 1. Comparison between proportional and linear response

3. Channel Coefficient

It is desirable that the data from the scintillation counter be represented in a form proportional to the initial gamma energy. The constant of proportionality is referred to as the channel coefficient and is represented by equation 1.

$$\text{Channel coefficient} = \frac{\text{incident gamma energy}}{\text{channel no.}} \qquad (1)$$

This equation can then be transposed to determine the energy range which a particular channel represents.

$$\gamma E = \text{C. C.} \times \text{channel no.}$$

where:

γE = incident gamma energy

C.C. = channel coefficient

This equation is of the form y = mx. It must be kept in mind therefore, that this calibration assumes a proportional response throughout the entire spectrometer system. This assumption is not entirely true.

B. Non-Linearity

As shown in Figure 2, a plot of pulse height vs energy is linear over most of the energy range but is not proportional since the intercept is not zero. There is also noticeable departure from linearity in the energy region below about .2 Mev.

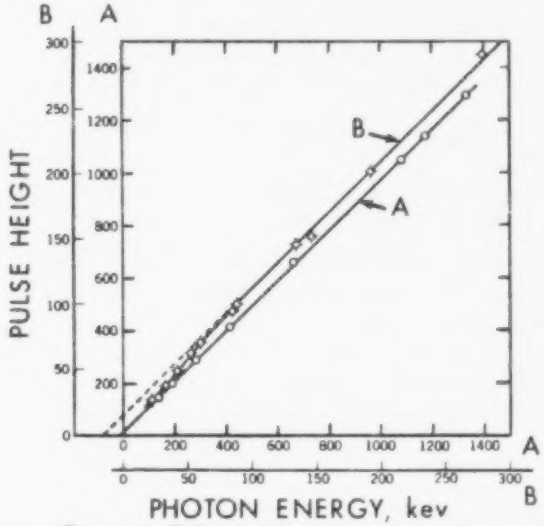


Figure 2. Pulse height versus energy for gamma rays on NaI(Tl) (ref. 1)

1. CAUSES OF NON-LINEARITY

There are two primary sources of non-linearity: the non-linear conversion of gamma energy into light photons, and the non-linear response of the linear amplifier. The former is prominent up to about .2 Mev, the latter is the primary contributor at high energies (>2 Mev).

C. Stability

Besides the problem of non-linearity, pulse height analyzers are not stable and tend to drift away from the position they were set for. There are two types of drift encountered. One is called zero shift which is a shift of the whole spectrum to the left or right. This type of shift results in an equal shift for all channels. Zero shift is almost always attributed to the analog to digital converter. The other is a drift in gain setting. This results in a proportional shift, channel for channel, depending on the pulse height represented by that channel. For example, a gain shift of 1 channel for a 0.1 Mev peak would correspond to a shift of 10 channels for a 1 Mev peak. This gain shift is caused by the detector and amplifier.

Some of the factors, which affect stability are count rate, warm up time, component conditions, temperature and magnetic field. When calibrating an instrument, one should be aware of both short term and long term drift.

III. CALIBRATION

As indicated, it is desired that the data from the scintillation spectrometer be displayed in proportional form with 0 energy falling in channel 0. It is well known that two points determine a straight line so all that is necessary to calibrate the counter is to establish the energy of two channels, remembering that E/channel no. = channel coefficient. This can be done by using two gamma ray sources, each emitting different energy gamma rays. When gamma sources are used, two peaks like the .662 Mev peak of Cs^{137} and the 1.17 Mev peak of Co^{60} are set into predetermined channels by manipulating the 0 control and the gain. The 0 control shifts the whole spectrum to the right or left and the gain control shifts or expands the spectrum. For instance, if the above mentioned peaks are set in channel 66.2 and channel 117 respectively, then the 0 has been set and the energy represented by a channel is .01 Mev times that channel number. Once the instrument has been calibrated, the zero setting is rarely changed. Minor shifts are corrected for by using the fine gain adjustment.

In preference to using only two points for calibration purposes, it is sometimes desirable to develop a complete calibration curve, i.e., gamma energy versus channel number, using a series of nuclides of varying energy. Although this procedure is more time consuming, and is not applicable as such, to a day by day procedure, it does yield more accurate results by taking into account the non-linearity of the system.

A. Choice of Channel Coefficient

No hard and fast rules can be said about choosing a channel coefficient although several guide lines may be kept in mind. Naturally an even number is desirable for easy handling mathematically. An excessively large channel coefficient will inhibit differentiation between peaks of similar energy. An excessively small channel coefficient enables good differentiation between peaks and is of particular value when examining the closely spaced peaks in the low energy range. It will cause the peaks themselves to be less distinctive however. This is particularly evident for high energy peaks. Further, using a small channel coefficient (large number of channels) may result in data handling problems.

An intermediate value of from .01 to .02 Mev per channel seems realistic for most work.

B. Determination of Peak Channel

One formidable problem encountered in energy calibration is accurately determining the exact, fractional peak channel of photopeak. Two methods may be used.

1. GRAPHICAL SOLUTION

This method merely entails plotting the uppermost points of a photopeak on an expanded linear scale. The points are connected by either straight lines or a smooth curve. A horizontal line is drawn at a height equal to 3/4 to 1/2 the peak activity. The midpoint of the horizontal line is considered to represent the peak channel (assuming a normal curve). This is shown in Example I.

EXAMPLE I

Data: Cs^{137} photopeak E = .662 Mev

Channel	cpm
30	14.5
31	34.5
32	63
33	67
34	50
35	23.5
36	8.5

a. Procedure

1. Data is plotted linearly as in Figure 3 (it should be pointed out that the more the plot is expanded, the more accurate are the results.)

2. A smooth curve (or straight line) is drawn between points.

3. A horizontal line is drawn at a height equal to 1/2 the maximum activity (line A-B).

4. The horizontal line is bisected.

5. The fractional peak channel equals the point of bisection.

Results:

peak channel = 32.77

2. MATHEMATICAL SOLUTION

If the photopeak is considered to represent a parabola (this turns out to be a reasonable assumption), a set of simultaneous

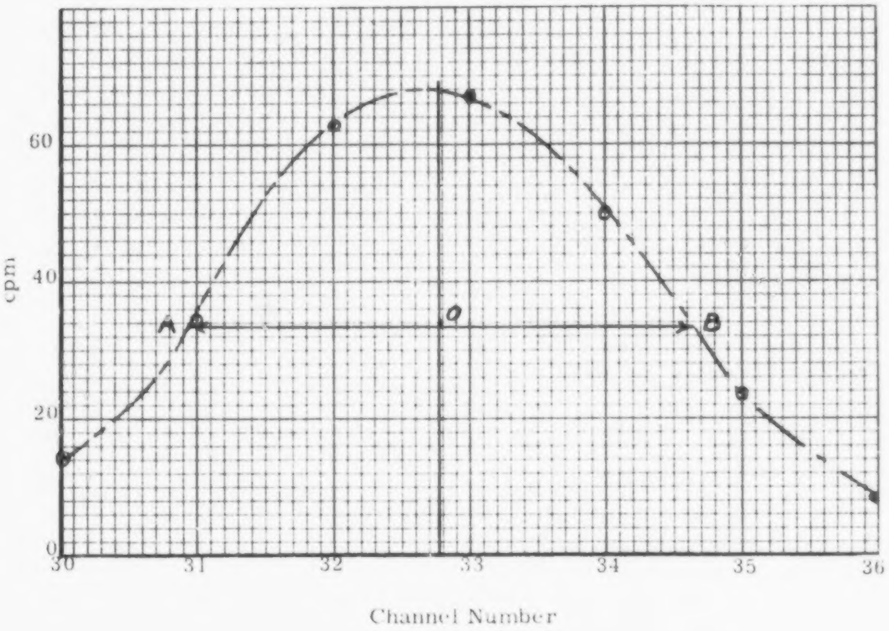


Figure 3. Graphical determination of fractional peak channel

equations may be set up utilizing the equation for a parabola. (Equation 4)

$$y = Ax^2 + Bx + C \qquad (4)$$

The peak channel will occur at

$\frac{dy}{dx} = 0$. This method is most easily demonstrated in Example II.

EXAMPLE II

Data: Cs^{137} photopeak E = .662 Mev

Channel 32	-	63 cpm
Channel 33	-	67 cpm
Channel 34	-	50 cpm

a. Procedure:

1. A parabola is assumed.

2. The constants in equation 4 are determined using simultaneous equations: (The actual values for the points may be utilized in the equations, or a shift in axis may be incorporated to simplify these numbers. Such a shift in axis is shown in Figure 4).

$$y_1 = Ax_1^2 + Bx_1 + C$$

$$y_2 = Ax_2^2 + Bx_2 + C$$

$$y_3 = Ax_3^2 + Bx_3 + C$$

$$13 = A(0)^2 + B(0) + C$$

$$17 = A(1)^2 + B(1) + C$$

$$0 = A(2)^2 + B(2) + C$$

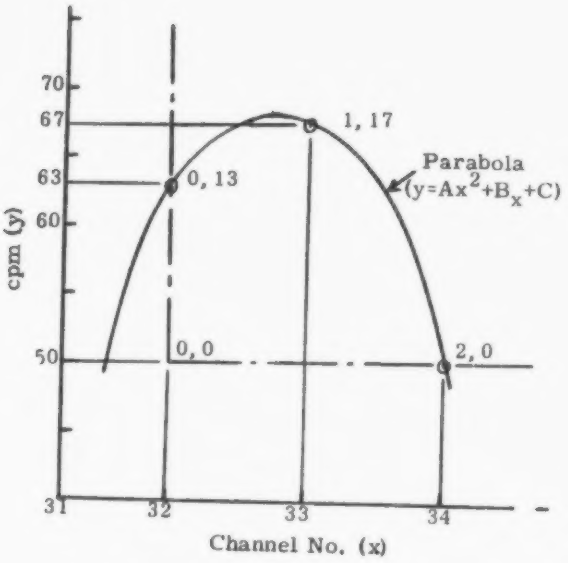


Figure 4. Mathematical determination of fractional peak channel

Values for constants from solution of equations:

$$C = 13, B = 14.5, A = -10.5$$

3. The constants are substituted into equation 4.

$$y = -10.5x^2 + 14.5x + 13$$

4. The equation is differentiated to determine $\frac{dy}{dx}$

$$\frac{dy}{dx} = 2(-10.5)x + 14.5$$

5. Peak channel is at $\frac{dy}{dx} = 0$

$$\frac{dy}{dx} = -21x + 14.5 = 0$$

$$x = \frac{14.5}{21} = .69$$

$$\boxed{\text{Peak Channel} = 32.69}$$

IV. SUMMARY

In order to realistically interpret a gamma spectrum it is necessary to correllate channel number to incident gamma energy. The constant of proportionality is called the channel coefficient and is given in terms of energy per channel number. This calibration assumes a proportional relationship between energy and channel number which is not always true. In addition a system is not always 100% stable. It is therefore necessary to recognize the presence of non-proportionality factors and instability factors when calibrating an instrument and minimize their effect where possible.

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GPO 886-756

Resolution

I. INTRODUCTION

The most indicative portion of a gamma spectrum for both qualitative and quantitative analysis is the characteristic photopeak. This peak displays a near Gaussian distribution and is generally described in terms of its resolution. Resolution is the peak width associated with the total absorption of monoenergetic radiation. It determines the ability of a system to distinguish between different energy gamma photons and thus is a critical factor effecting the accuracy of spectral analysis.

II. DEFINITION

The common definition of resolution is given in terms of percent resolution, as shown in equation (1).

$$\text{Percent Resolution } (\% R) = \frac{\Delta E_{\frac{1}{2}} \times 100}{E} \qquad (1)$$

where: E = photopeak energy (Mev)

$\Delta E_{\frac{1}{2}}$ = energy interval (Mev) between points at one-half of the maximum peak height

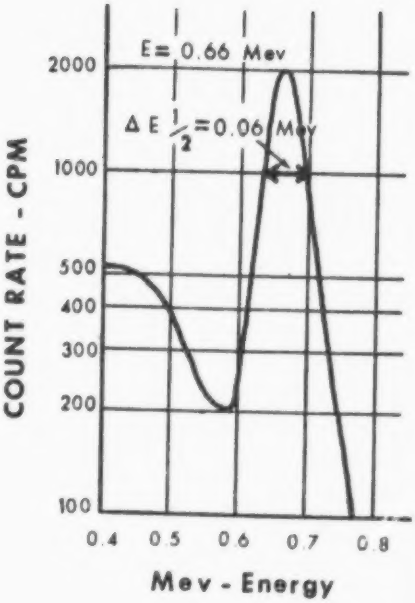
An example of the determination of percent resolution is as follows (See Figure 1).

Example:

E = 0.66 Mev

Photopeak height = 2000 cpm

½ Photopeak height = 1000 cpm



DETERMINATION OF % RESOLUTION
Figure 1

$$\Delta E_{\frac{1}{2}} = .70 - .64 = 0.06$$

$$\% R = \frac{0.06 \times 100}{.66} = 9.1\%$$

Because the photopeak follows a near Gaussian distribution, the energy interval ($\Delta E_{\frac{1}{2}}$ can be related to the standard deviation by a constant factor as given in equation (2).(1)

$$\Delta E_{\frac{1}{2}} = 2\sqrt{2 \ln 2}\,\sigma$$

$$\Delta E_{\frac{1}{2}} = 2.36\,\sigma \qquad (2)$$

where: σ = standard deviation of the Gaussian distribution

III. FACTORS AFFECTING RESOLUTION

The fact that the total absorption of monoenergetic gamma photons produces a broad photopeak rather than a line spectrum results from the statistical nature of the processes involved in the detector system and the nonproportional response of the crystal itself.

A. Statistical Variation

The statistical variation of the detector response is caused by several separate but inter-related factors.

1. The number of photons emitted per scintillation event in the crystal. (Phosphor fluorescent efficiency.)
2. The fraction of the light originating in the scintillation which ultimately reaches the photocathode. (Light collection efficiency.)
3. The fraction of photons that are converted to electrons at the photocathode. (Photocathode efficiency and uniformity.)
4. The fraction of photoelectrons collected by the first dynode. (Photoelectron collection efficiency.)
5. The secondary emission ratio at the succeeding dynodes.

The first two items are associated with the characteristics of the crystal itself. Their combined effect can be designated as a constant α.(3)

The last three items are functions of the photocathode and photomultiplier. Variance of these factors is known to be statistical in nature and dependent on energy. The higher the energy of an incoming photon the more light is produced in the crystal and subsequently the more photoelectrons are produced at the photocathode. From poisson statistics, which hold in this case, the variance, in terms of the standard deviation is:

$$\sigma = \sqrt{N}$$

where:

N = average number of electrons

It is seen that as N increases, the standard deviation and thus $\Delta E_{\frac{1}{2}}$ increases as the square rrot of N. The combined effect of the statistical variations of the photomultiplier response can be expressed as:

$$\frac{\beta}{E}$$

The total % resolution can now be represented by the sum of the effects of α and β.(3)

$$R^2 = \left(\frac{\Delta E_{\frac{1}{2}}}{E}\right)^2 = \alpha + \frac{\beta}{E} \qquad (3)$$

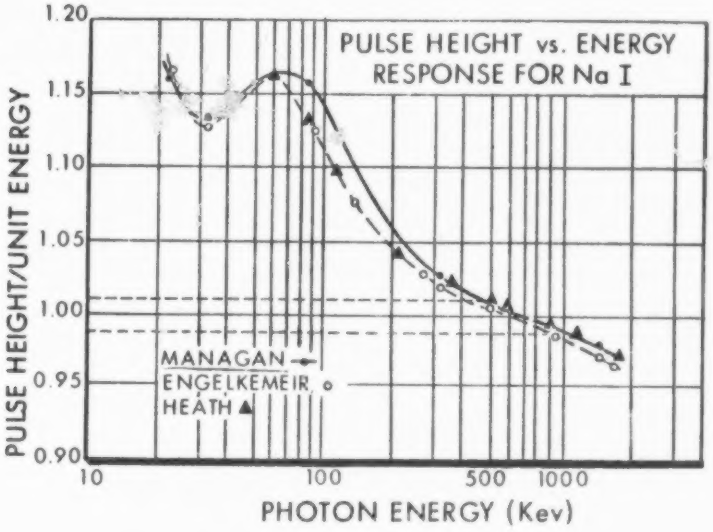
or

$$(\%R)^2 = \left(\frac{\Delta E_{\frac{1}{2}}}{E} \times 100\right)^2 = \left(\alpha + \frac{\beta}{E}\right)(10^4) \qquad (4)$$

This relationship is shown as the solid line in Figure 3. It is important to note that this formula assumes a Gaussian distribution of light photons produced by monoenergetic incident rays, with no dependence of α on energy. This assumption does not always hold true.

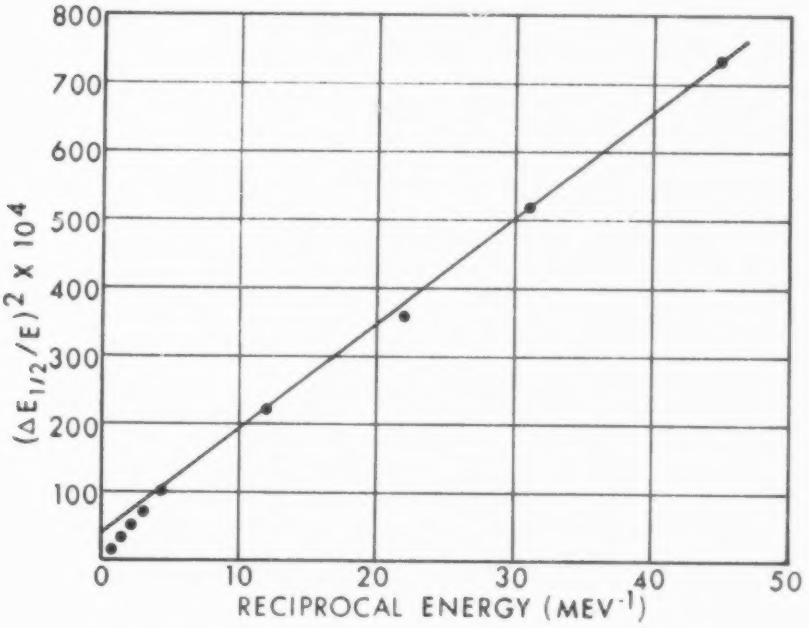
B. Non-Proportionality of Crystal Response

The non-proportional response of the NaI(T1) crystal to gamma photons, (Figure 2,) exhibits a significant influence on resolution as indicated by the deviation of $\frac{\Delta E_{\frac{1}{2}}}{E}$ from the theoretical curve at low values of $\frac{1}{E}$ in Figure 3. A total % resolution of 7.7% for a .66 Mev photopeak corresponds to the com-



NON-PROPORTIONALITY OF NaI(Tl) (REF. 1)

Figure 2



% RESOLUTION VS. RECIPROCAL ENERGY (REF. 2)

Figure 3

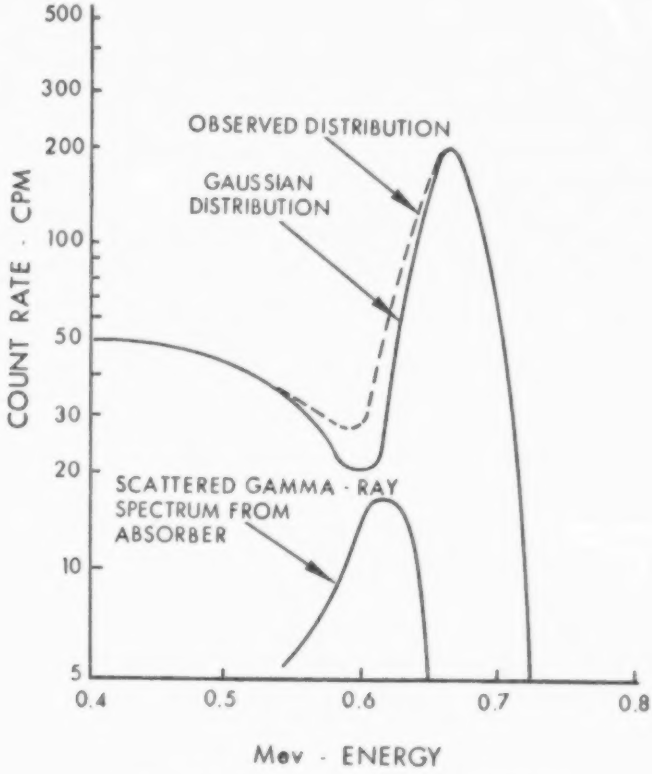
bined effect of a 4% resolution resulting from statistical variation in the photomultiplier and a 6.6% resolution resulting from intrinsic crystal resolution.(3) It further turns out that variations in the uniformity of the photocathode have little effect on this resolution. This shows that in this case the non-proportional response of the crystal to gamma energy is the major contribution. It has been found that in the energy range of .046 to 1.33 Mev the scintillation efficiency, i.e., pulse height per unit energy, varies by about 20%.(4) This can be seen in Figure 2.

The net effect of gamma energy on % resolution is shown in Figure 3. It is important to realize that although the peak width ($\Delta E_{\frac{1}{2}}$) increases as about the square root of increasing gamma energy, the % resolution $\left(\frac{\Delta E_{\frac{1}{2}}}{E} \times 100\right)$ by definition decreases.

C. Compton Interference

If one combines the effects of photomultiplier statistics and crystal non-linearity, a photopeak representing a near Gaussian distribution results.(5) In actual practice however, the photopeak deviates from a Gaussian shape and appears skewed to the low energy side. (See Figure 4.)

In order to account for this phenomena a third resolution factor must be introduced. This skewness is the result of scattered gamma rays from the crystal can or beta



TYPICAL PHOTOPEAK DISTRIBUTION

Figure 4

absorber.(5) This small angle backscatter not only results in a skewing of the photopeak, and increasing the resolution, but also accounts for the filling up of the Compton valley.

IV. SUMMARY

A detailed analysis of all the individual factors which influence resolution becomes rather complex to say the least. In general, however, two things can be said. First, resolution is primarily the net result of the non-proportional response of crystals to gamma rays, and the statistical variation of the photomultiplier (including the photocathode). Second, % resolution displays a net inverse relationship to gamma energy.

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GPO 886-557

Evaluation of Linearity and Stability

I. INTRODUCTION

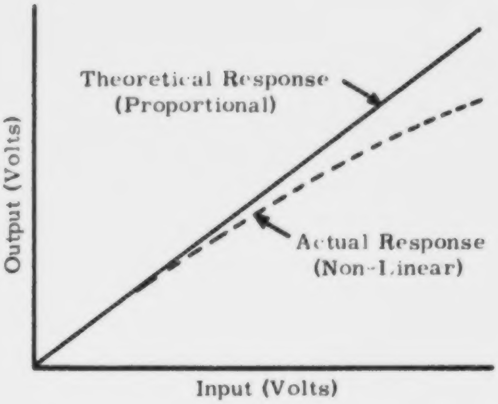
It is recognized that the pulses being processed through a gamma spectrometer do not always maintain a proportional or even a linear correlation to incident gamma photon energy. It is further recognized that a spectrometer system may not be stable, i.e., shifts in gain setting and zero setting may occur. It therefore becomes essential to be able to detect, evaluate, and compensate for such irregularities in order to realistically and accurately interpret data.

II. NON-LINEARITY

A Causes of Non-Linearity

There are two main sources of non-linearity. The first is the non-linear conversion of gamma energy into light photons in the crystal. The scintillation efficiency (amount of light produced per unit energy) is not constant but is a function of energy. This non-linearity is prominent up to about .2 Mev after which it is usually considered negligible.(1)

The second source of non-linearity is in the analyzer portion of the spectrometer (the amplifiers, analog to digital converter, and associated circuits). The most important of these is the linear amplifier. The purpose of the amplifier is to linearly multiply the input pulse (from the detector) to a larger output pulse capable of analysis in the pulse sorter mechanism. The gain or multiplication factor may not be constant over the entire rated output range of the amplifier however.(1) This is shown in Figure 1.



Non-Linear gain of linear amplifier

Figure 1

A similar non-linear response can be present in the analog to digital converter. The analyzer as a whole is the primary contributor to non-linearity at high energies.

The net effect of non-linearity, whether it be from the detector or the analyzer, is twofold. First, it distorts the energy axis and second, it causes either an excess or a deficiency of counts in that region as compared to the same region displaying linear gain.(1)

B. Definitions of Linearity

Linearity is generally expressed in one of three ways.

1. Absolute linearity is defined as the ratio of the deviation in energy as determined from the channel coefficient, to the true

energy. With reference to Figure 2, absolute linearity may be expressed as:

$$\text{Absolute linearity} = \frac{\Delta E}{E\gamma} \qquad (1)$$

where:

E_γ = actual γ energy

E_o = energy observed

ΔE = deviation from true energy
$= E_\gamma - E_o$

It is evident that linearity, so defined, is directly dependent on E_γ. In other words, for a given energy deviation (ΔE), several values of absolute linearity may be determined depending on the reference energy used (E_γ). For this reason absolute linearity is not a readily interpretable parameter unless the reference energy is specified.

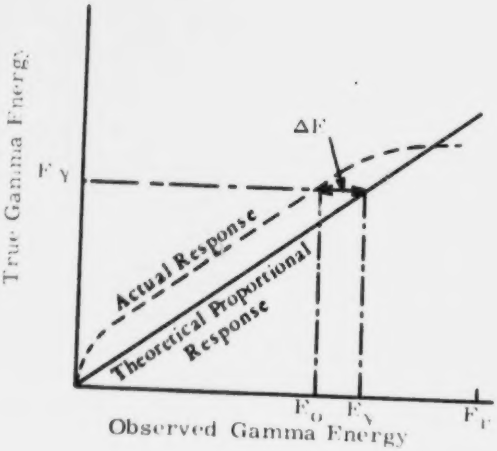
2. Integral Linearity is expressed as the ratio of the error in energy to the full energy scale. From Figure 2 this would be:

$$\text{Int. Lin.} = \frac{\Delta E}{E_r} \qquad (2)$$

where:

E_r = maximum rated energy = channel coefficient × maximum channel number.

ΔE generally refers to the maximum deviation from proportionality. Integral linearity is a commonly used term but it has its limitations. The ratio of the error in energy to the true energy (absolute linearity) may not be good in the low energy region but the integral linearity will not reveal this. Also, the choice of E_r is not fixed, in that it is determined by the energy range of interest.



Effect of non-linearity on spectrometer response
Figure 2

3. Differential Linearity is a measure of the uniformity of the channel widths. For instance a differential linearity of 2% means that no channel departs from the average channel width by more than 2%.

C. Measurement of Non-Linearity

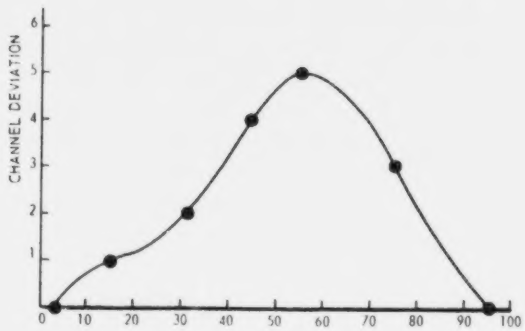
A measurement of absolute or integral linearity can be made in one of two ways.

1. USING STANDARD SOURCES

A graph of photopeak channel vs energy is made for a series of radionuclides of different known gamma energies. The deviation from the normal (linear) curve can then be ascertained. This is shown in Figure 2. This method depicts the linearity of the entire system (i.e., detector plus analyzer). Often it is more desirable to plot channel deviation vs photopeak channel. This method yields a more sensitive representation as shown in Figure 3.

2. USING PULSE GENERATOR

A pulse generator may be substituted for the series of radionuclides and channel no. vs energy plotted as before. This method



Channel number typical channel Deviation vs channel number curve

Figure 3

provides a check on the analyzer system alone since the detector system is eliminated.

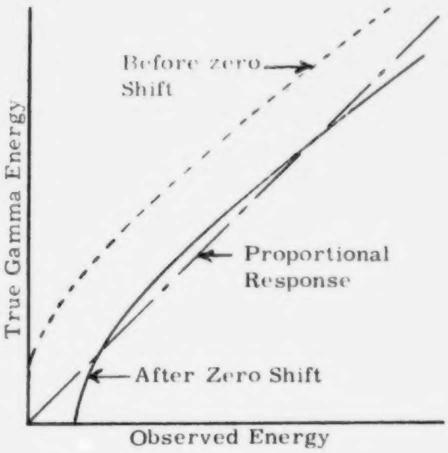
Both the linearity of the total system and of the analyzer alone should be known. A total maximum integral linearity of .2% is desirable for a 200 channel range.

The measurement of differential linearity can be accomplished with a sweeping pulse generator.(4) The pulses, increasing slowly in amplitude, are applied to the analyzer. These will be tallied in successively higher channels. If the pulse amplitudes are increased linearly in time and the frequency is constant, the number of pulses tallied in each channel is the measure of the channel width. A differential linearity of 1-2% is usually desirable.

D. Corrections for non-linearity

The non-linearity of the scintillation counting system is an intrinsic part of the equipment and cannot therefore be eliminated. Of course, the use of amplifiers and analyzers of superior design and performance will minimize this effect. In some analyzers the amplifiers actually are designed to compensate in part for the non-linear response of the scintillation crystal.

One course of action that can be taken to help minimize the effect of non-linearity is incorporating zero offset. This merely consists of shifting the zero channel so that it represents some energy higher or lower than zero. The effect of this shift is to make the spectrometer response more nearly proportional over a given energy range. This is shown in Figure 4.



Effect of zero shift on non-linearity

Figure 4

It is seen that a linear response can be made proportional. A non-linear response can only be shifted to correspond, on an average, to a proportional response.

III. STABILITY

Besides the problem of non-linearity, pulse height analyzers are not stable and tend to drift away from the calibration they were set for. There are two types of drift encountered. One is called zero shift which is a shift of the whole spectrum to the left or right. This type of shift results in an equal shift for all channels. Zero shift is almost always attributed to the analog to digital converter. The other is a drift in gain setting. This results in a proportional shift, channel for channel, depending on the pulse height represented by the channel. For example, a gain shift of 1 channel for a 0.1 Mev peak would correspond to a shift of 10 channels for a 1 Mev peak. This gain shift is caused by the detector and amplifier with the photomultiplier being the primary contributor. In general gain shift represents the most critical problem. Most of the new analyzers are fairly stable against zero shift.

A. Causes of Instability

Some of the factors which affect instrument stability are: warm-up time, component condition, count rate, temperature, and the earths magnetic field. These factors influence the secondary emission ratio of the dynode surfaces of the photomultiplier tube which, as indicated above, is the primary contributor to gain shift.

B. Measurement of Stability

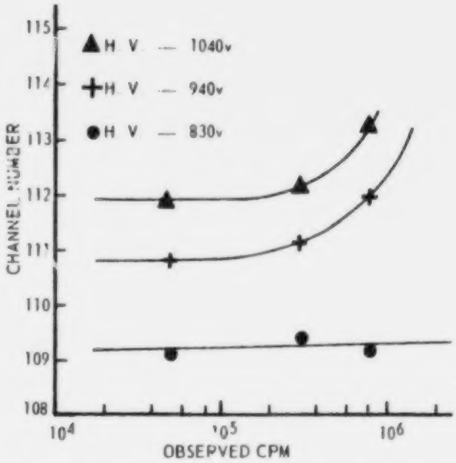
The test for gain shift or zero shift with time is merely to frequently check the position of the photopeak of a standard source or even better, the relative positions of two photopeaks. Shift with activity can be checked by observing the position of the peak of a single standard at different levels of activity.

C. Correction for Instability

There are a number of principles which may be employed to correct for or minimize the effect of instability.

1. It is advisable to operate the photomultiplier at a minimum feasible high voltage and use maximum amplifier gain. This is because the phototube is more sensitive to gain shift than the amplifier. The effect of using a low high voltage setting is depicted in Figure 5.(3)

In this figure, channel shift is shown as a function of increased sample activity and H. V. setting. The total gain was kept fairly constant by increasing the amplifier gain as the high voltage was decreased. It is noted that for the same total gain, less gain shift resulted with increased sample activity at the lower voltage settings.

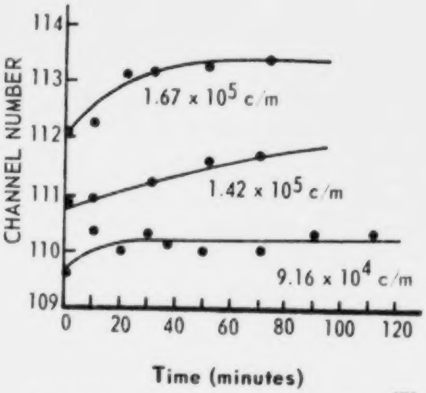


Effect of counting rate on gain at various photomultiplier tube voltages(3)

Figure 5

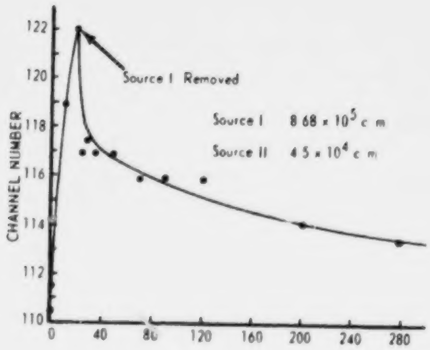
2. Do not count high activity samples any longer than necessary. Any method of reducing the count rate from high activity samples such as, diluting the sample, decreasing the geometry or incorporating absorbers, may be beneficial. Figure 6 depicts gain shift with time for samples of varying activity. Less shift is noted for the samples of lower activity.

Another factor to be taken into consideration is recovery time. The effect of a pulse height shift caused by a high activity sample



Long-term counting rate gain shifts(3)

Figure 6



Time (minutes) gain recovery vs time after replacing a high activity sample with a low activity sample

Figure 7

may remain for some time after the sample is removed and influence the counting of subsequent samples. This effect can be seen in Figure 7. In this figure, a high activity sample was counted for thirty minutes and then replaced by a low activity sample.

3. Frequently check the instrument calibration with a standard source such as Cs^{137}. This should be done before and after all counts.

4. Maintain instrument at a constant temperature.

5. Gain stabilization devices or normalization of data may be incorporated to compensate for gain shift.

a. Gain stabilization devices are generally based on an electronic feedback principle. This usually entails the introduction of standard pulses into the instrument. This may be accomplished by incorporating an alpha source within the crystal, gamma sources exterior to the crystal, or with light generators within the detector. The resulting pulse may then be amplified along with the normal pulses and fed back through a feedback circuit which in turn controls the high voltage to the photomultiplier. Any change in the height of the standard pulse will cause a corrective change in the high voltage setting. This method provides a continuous correction for gain shift with time during a counting period.

b. Normalization of Data. This technique is a mathematical one applicable to computors. The photopeak of the sample spectrum is shifted by extrapolation to fit a specified calibration - that of a standard. It is important to note that this procedure does not correct for gain shift occurring during the counting period. It only shifts the resulting spectrum to some predetermined calibration and zero setting.

IV. SUMMARY

The energy calibration of a gamma spectrometer and subsequnet interpretation of its data assumes a proportional relationship between energy and channel number. This assumption may be in error. In addition, a system is not always 100% stable. It is therefore necessary to recognize the presence of non-proportionality factors and instability factors in an instrument and minimize their effect wherever possible.

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GPO 886-417

Counting Efficiency of Gamma Spectrometers

I. INTRODUCTION

A knowledge of detector efficiency of a gamma spectrometer and the variables which effect it, is necessary for at least three reasons.

A. An accurate numerical value is required to convert the count rate, obtained from spectral analysis, to units of activity.

B. Efficiency will change with physical changes of the counting system and the environment surrounding it. It is undesirable to have changes occur under uncontrolled conditions.

C. For low level counting of environmental samples, it is desirable to increase the efficiency as much as possible to increase the minimum level of detectability.

This outline will define and discuss efficiency, variables that affect efficiency, and commonly used counting arrangements.

II. COUNTING EFFICIENCIES

Total efficiency is defined as the ratio of the observed count rate in a selected number of channels, to the true disintegration rate of the specific radionuclide which calibration is being measured.

$$\text{Total Efficiency} = \frac{\text{counts per minute}}{\text{disintegrations per minute}}$$

In some types of gamma spectroscopy, gamma efficiency is used. Gamma efficiency is defined as the ratio of the observed count rate to the gamma emission rate which is defined as the product of the disintegration rate and gamma abundance.

$$\text{Gamma Efficiency} = \frac{\text{cpm}}{\gamma\text{pm}} = \frac{\text{cpm}}{\text{dpm} \times \gamma\text{ abundance}}$$

Efficiency can be determined by comparative counting (using a standard source and observing a count rate) or by absolute counting (calculating the individual factors involved in a counting system). The latter method is quite involved but, when done by experienced personnel can be very accurate.

In comparative determination of efficiency, it is necessary to reproduce the counting arrangement, and conditions of the standard and the sample. Therefore, a separate counting efficiency must be determined for each nuclide, counting arrangement, and portion of the spectra being calibrated.

An illustration of determining total efficiency by comparative counting for different portions of the spectra, Figure 1, is given in Example 1.

Example 1

Required:

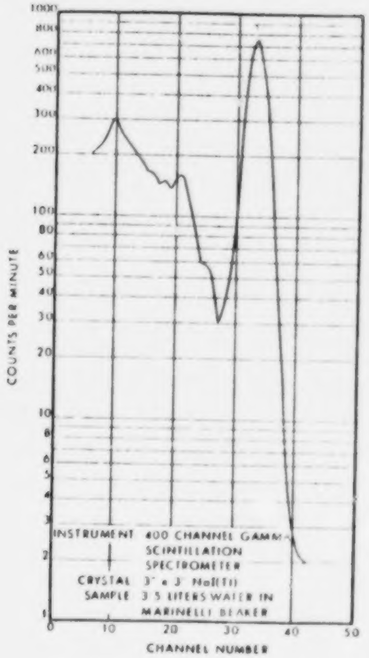
a) Determine total efficiency for
    1) Gross Spectrum $_{(1-50)}$
    2) Peak Channel $_{(33)}$
    3) Photopeak Channels $_{(30-36)}$

b) Determine gamma efficiency for peak channel$_{(33)}$

Data:

Nuclide: Cs^{137}

Counting Arrangement: Marinelli beaker



GAMMA SPECTRAL DATA FOR FIGURE 1

CHANNEL NO.	COUNTS PER MINUTE
05	[illegible]
06	[illegible]
07	[illegible]
08	[illegible]
09	[illegible]
10	[illegible]
11	[illegible]
12	[illegible]
13	204
14	188
15	[illegible]
16	165
17	144
18	[illegible]
19	[illegible]
20	[illegible]
21	[illegible]
22	[illegible]
23	[illegible]
24	[illegible]
25	[illegible]
26	[illegible]
27	[illegible]
28	[illegible]
29	[illegible]
30	[illegible]
31	[illegible]
32	[illegible]
33	[illegible]
34	[illegible]
35	[illegible]
36	[illegible]
37	[illegible]
38	[illegible]
39	[illegible]
40	[illegible]

GAMMA SPECTRA FOR Cs^{137}

Figure 1

Volume: 3.5 liters
Spectra: See Figure 1
Added activity: 1.4×10^5 dpm
Detector: Rhino I
Energy calibration: 20 Kev/channel
Time of Count: 5 min.
Solutions:

a) 1. Sum of channels$_{(-50)}$ = 32880

$$\text{cpm} = \frac{32880}{5} = 6576$$

$$\text{Total Eff.}_{(\text{gross})} = \frac{\text{cpm}}{\text{dpm}} = \frac{6576}{1.4 \times 10^5} = 0.047$$

2. Sum of channel$_{(33)}$ = 3792

$$\text{cpm} = \frac{3792}{5} = 758$$

$$\text{Total Eff.}_{(\text{peak})} = \frac{\text{cpm}}{\text{dpm}} = \frac{758}{1.4 \times 10^5} = 0.0054$$

3. Sum of channels$_{(30-36)}$ = 14010

$$\text{cpm} = \frac{14010}{5} = 2802$$

$$\text{Total Eff.}_{(\text{photopeak})} = \frac{\text{cpm}}{\text{dpm}} = \frac{2802}{1.4 \times 10^5}$$

$$= 0.02$$

b) Gamma Efficiency

cpm = 758

$$\text{Gamma Eff.}_{(\text{peak})} = \frac{\text{cpm}}{\text{dpm} \times \text{gamma abundance}}$$

$$= \frac{758}{1.4 \times 10^5 \times 0.823} = 0.00658$$

An application of efficiency calculations is best shown by Example 2.

Example 2

Required: Calculate the activity of the following sample

Data:

Volume: 0.5 liter
Net Count: 60 cpm
Total Eff.$_{\text{(photopeak)}}$ = 0.02

Solution:

$$\text{pc/l} = \frac{\text{cpm}}{\text{Total Eff.} \times 2.22 \text{ dpm/pc} \times \text{volume}}$$

$$= \frac{60}{0.02 \times 2.22 \times 0.5}$$

$$= 2700 \text{ pc/l}$$

III. EFFICIENCY FACTORS

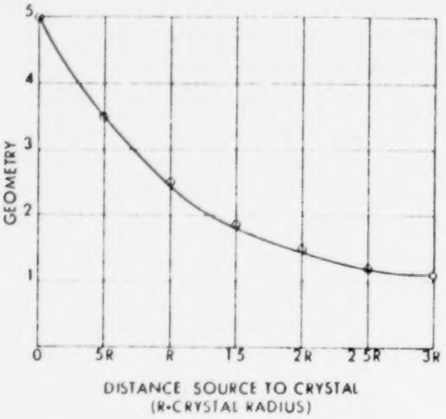
The total efficiency is actually made up of two individual factors: geometry and incident intrinsic efficiency. A change in either of these will change the total efficiency.

A. Geometry

Geometry is defined as that percent, Ω, of the solid angle, 4π, about a source which is subtended by the sensitive volume of the detector.(1) With this definition in mind, let us consider some of the parameters, i.e., distance, crystal shape, sample container and size, that will effect the geometry.

1. DISTANCE: SOURCE TO CRYSTAL FACE

From the definition of geometry it can be seen that the best geometry obtainable with a solid crystal is when a point source is in contact with the crystal face. In this arrangement half of the emitted rays are entering the crystal. If all the rays that are emitted into the crystal cause a measureable scintillation, then we can expect a total gross efficiency of 50 percent. As the source is moved further away from the crystal, fewer and fewer of the total emitted rays will be entering the crystal and the efficiency will decrease. Figure 2 illustrates the effect of distance on geometry. It should be noted that the source was moved on a line through the axis of the crystal.



GEOMETRY OF CRYSTAL AS A FUNCTION OF DISTANCE FROM SOURCE TO CRYSTAL FACE

Figure 2

2. CRYSTAL SHAPE

As previously noted, for any solid right cylindrical crystal, the maximum geometry obtainable is 50 percent.

In order to increase the geometry, a well type crystal may be used for small sources. A well crystal is a solid right cylindrical crystal with a hole drilled part way into the center. The effect is that the percent of the solid angle subtended is greatly increased. A 3" × 3" crystal with a well 1/2" diameter × 1 1/2" deep will give a geometry of 97 percent.(4) An immediate disadvantage of a well crystal is the sample volume which is usually limited to about 10 ml.

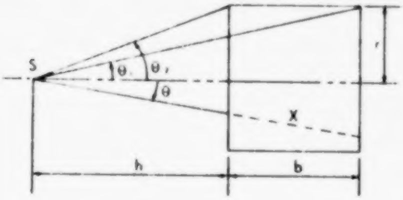
Another type of crystal has a hole drilled through the longitudinal cross section and fitted with concentric tubing so radioactive materials may be passed through the tubing to obtain geometric efficiency close to 100 percent.(4) Again the sample volume imposes a limitation on the use of this crystal. Also decontamination is a serious problem if the crystal should become contaminated.

3. SAMPLE CONTAINER

In environmental counting the sample does not take the form of a point source but is more normally distributed as a plane surface or contained as a volume, above or around the crystal. As disintegrations take place everywhere within the sample, i.e., at any distance from the crystal, the efficiency determined for a nonpoint source is actually an integrated efficiency of each point within the sample. The highest counting efficiency for a given volume is when the average distance from the source to the crystal is a minimum. Therefore, a sample contained in a 3" × 3" cylindrical volume will give a higher counting efficiency than the same sample contained in a 1" × 27" cylindrical volume.

B. Incident Intrinsic Efficiency

The intrinsic efficiency is more difficult to evaluate numerically than the geometry factor. The incident intrinsic efficiency, $E_{(E)}$, is defined as the fraction of the monochromatic isotropic radiation with photon energy (E), incident on the crystal face, which interacts to produce a measureable scintillation.(2) The probability of interaction of a photon incident on the detector face will be different with the placement of the source. The probability that a photon traversing a path x, see Figure 3, in the crystal with no interaction is $e^{-\mu x}$ where μ is defined as the linear attenuation coefficient in cm^{-1} for a particular crystal and a monoenergic photon. The probability that there will be an interaction is then $(1 - e^{-\mu x})$. Therefore, for a collimated source the incident intrinsic efficiency is $E_{(E)} = (1 - e^{-\mu x})$.



SCHEMATIC DIAGRAM OF SOURCE

Figure 3

For a non-collimated point source, a distance h from the crystal, there would be rays impinging at different angles which would give different path lengths, x. In order to find the mean path length, we would have to integrate the path lengths over the entire crystal. In previously defined terms:

$$E_{(E)} = \frac{1}{\Omega_o} \int (1 - e^{-\mu x})\, d\Omega$$

where x is now a function of θ, b, and h which are defined by Figure 3; and μ which is defined as the attenuation coefficient.

The solid angle, $4\pi\,\Omega_o$, is the solid angle subtended by the crystal at the point source and is a function of h and r or of θ_2 alone. It follows that

$$4\pi\,\Omega_o = 2\pi \int_o^{\theta_2} \sin\ \theta d\theta$$

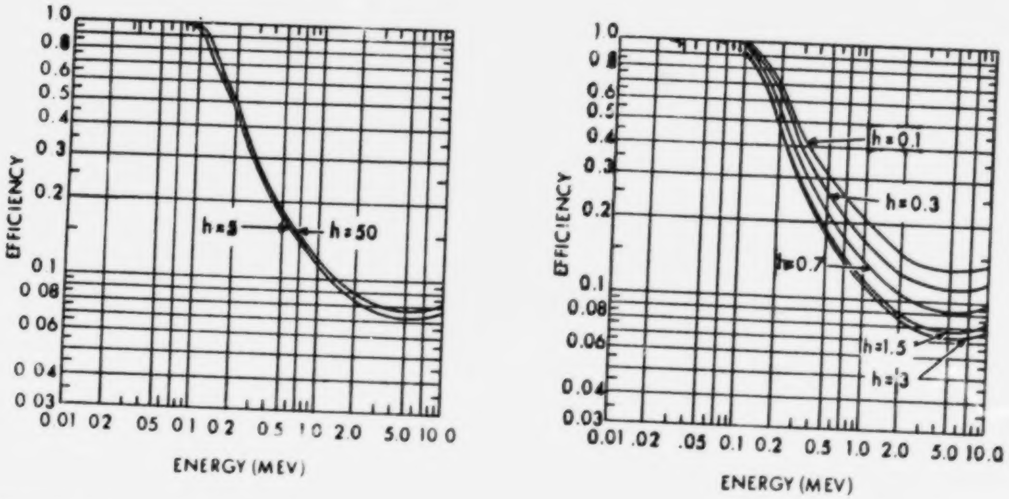
then

$$E_{(E)} = \frac{1}{2\,\Omega_o} \int_o^{\theta_2} (1-e^{-\mu x}) \sin \theta d\theta$$

Figure 4 illustrates the incident intrinsic efficiency versus energy for sources varying in distance, h. It should be noted that the intrinsic efficiency decreases with increasing distance until the minimum average path is reached and then increases as the interacting rays become more collimated. The calculations for the curves in Figure 4 were performed on a IBM type 650 computer.

1. CRYSTAL SIZE

Because the path lengths, for the same gamma energy, will increase, higher efficiencies will result from larger crystals. An example of this would be the comparison of a 1" × 1" NaI(Tl) crystal with a 3" × 3" NaI(Tl) crystal.



INCIDENT INTRINSIC EFFICIENCY VS. ENERGY FOR 1.5 in. DIAMETER NaI(Tl) CRYSTAL

Figure 4

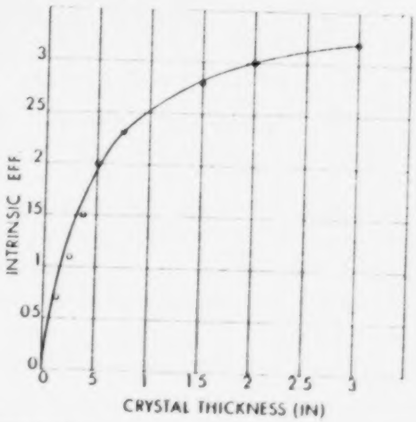
If we had a 0.5 Mev gamma energy source we could expect about 22 percent and 45 percent intrinsic efficiencies, respectively. Therefore, if higher intrinsic efficiencies are desired, larger crystals must be used (see Figure 5). At the present time there is a limit to the size of the crystals due to the process by which they are made. Economic consideration must also be taken into account as larger crystals are very expensive in relation to the efficiency gained.

2. EFFECTS OF ENERGY ON EFFICIENCY

Equally important in determining efficiency is the energy of the gamma ray being measured. Gamma rays are electromagnetic radiations or photons. The probability of gamma interaction is thus inversely proportional to the γ energy up to about 5 Mev and also varies with the mode of interaction. Figure 6 illustrates the effect of energy and mode of interaction on efficiency in terms of cross section.

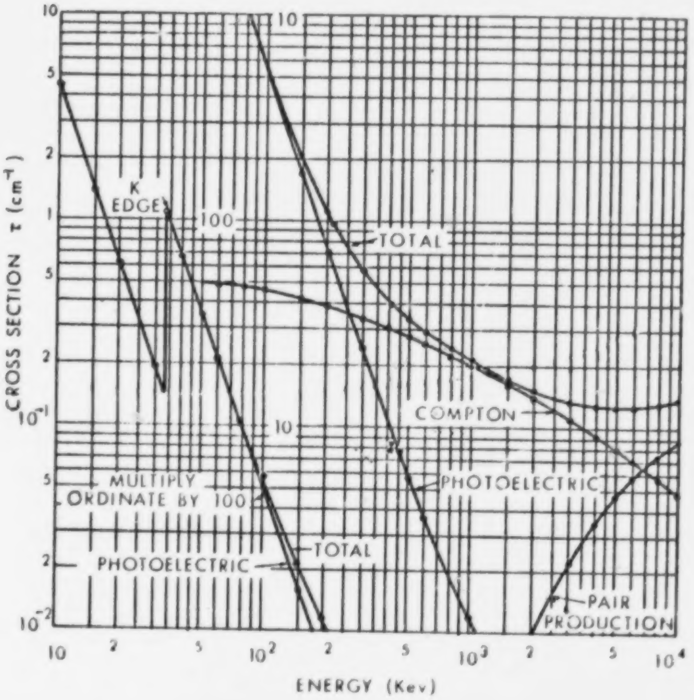
3. EFFECTS OF TEMPERATURE ON EFFICIENCY

At the present time there has not been sufficient research done on the effects of temperature on efficiency. The main problem is to hold the instrumentation and phototubes at a constant temperature while varying the temperature of the crystal. From the work that has been done it appears that a higher efficiency is obtained for lower temperatures. The range of temperatures, where the most change in efficiency was noted, was



INTRINSIC EFFICIENCY VS. CRYSTAL THICKNESS FOR A 1 MEV POINT SOURCE LOCATED 2 CM FROM A 1.5" DIAM. CRYSTAL

Figure 5



TOTAL NaI ABSORPTION CURVE FOR GAMMA INTERACTION

Figure 6

substantially below room temperature. There appears to be little change in efficiency in the range of 65° F. to 80° F. which would normally be the range of room temperature variation. However, counting of frozen samples, thereby lowering the temperature of the crystal, may have significant effects on the efficiency.

IV. SUMMARY

The efficiency of a scintillation counter arrangement depends on various characteristics of the system and the sample. Efficiencies must be known in order to compute the activity from a count rate. Most of the variables that go to make up the efficiency can be controlled by the operator. The most important parameters are the crystal size, type and composition, the geometrical arrangement, and the energy of the gamma rays which are being studied. These variables have a pronounced effect on the total efficiency.

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Nuclear Counting Statistics

I. INTRODUCTION

This lecture has a two-fold purpose: (1) to convey fundamental concepts of the statistical nature of radioactive decay, and (2) to explain and illustrate methods of using this characteristic to the best advantage in laboratory counting.

II. THE STATISTICAL NATURE OF RADIOACTIVE DECAY

The law of radioactive decay is a statistical law and is the result of a very large number of events subject to the laws of probability. There is some average number of atoms which disintegrate in one second, but the number which break up in any given second shows fluctuations around this value. The magnitude of these fluctuations can be calculated with the aid of the theory of probability, and the results provide a measure of the precision or trustworthiness of data obtained in the laboratory.

The exponential law of decay was deduced by von Schweidler in 1905 in a quite general form, without any special hypothesis about the structure of radioactive atoms or about the mechanism of disintegration. He assumed only that (1) the disintegration of an atom of a radioactive element is subject to the laws of chance, and that (2) the probability P for an atom to disintegrate in a time interval Δt is independent of the past history of the atom and (3) is the same for all atoms of the same type. The probability of disintegration then depends only on the length of the time interval and, for sufficiently short intervals, is proportional to Δt. Then $P = \lambda \Delta t$, where λ is this constant of proportionality, which has come to be known as the disintegration constant of the substance. The probability that the given atom will last (i.e. NOT disintegrate) during the short interval Δt is: $(1 - P) = (1 - \lambda \Delta t)$. If the atom survives this interval, then the probability that it will survive a second time interval Δt is again $(1 - \lambda \Delta t)$. The probability that the given atom will survive both the first and second intervals is $(1 - \lambda \Delta t)^2$; for n such intervals, the probability of survival is $(1 - \lambda \Delta t)^n$. If the total time $n(\lambda t)$ is set equal to t, then the probability of survival for the total time t is $(1 - \lambda \frac{t}{n})^n$. The probability that the atom will remain unchanged after any time t is the limit of the above probability as Δt becomes vanishingly small, or as n becomes very large. In the calculus a definition of the exponential function is derived:

$$e^{-x} = \lim_{n \leftarrow \infty} (1 - \frac{x}{n})^n$$

from which it follows that:

$$\lim_{n \leftarrow \infty} (1 - \frac{\lambda t}{n})^n = e^{-\lambda t}$$

The statistical interpretation of this result is that if there is initially a large number N_o of radioactive atoms, then the fraction remaining unchanged after time t is:

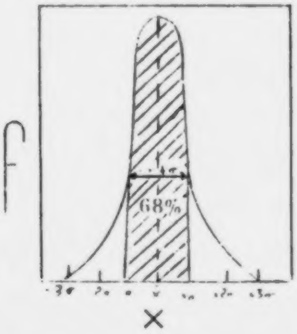
$$\frac{N}{N_o} = e^{-\lambda t}$$

where N is the number surviving to time t.

III. THE APPLICATION OF STATISTICAL TECHNIQUES TO COUNTING

Radioactive disintegrations occur in a manner described by the Poisson (pronounced PWAH - SOHn) distribution, which gives the frequency of observation of the number of counts n for a true or average number ν. For a moderately large number of events (≥ 30), the so-called "normal curve" or Gaussian distribution whose mean is ν and whose standard deviation is $(\nu)^{\frac{1}{2}}$ is a good approximation to the Poisson distribution. Because it is less complex to work with, the normal distribution is generally used in counting statistics instead of the Poisson distribution.

The "normal" or "bell-shaped" curve is obtained by plotting the frequency of observation of the quantity x against the numerical value measured for the quantity x. The curve is determined by two parameters, the mean μ or $\bar{x}$, and the standard deviation σ. The maximum value of observed frequency f occurs at the mean value of x, i.e. $\bar{x}$. The curve is broad if σ is large and narrow if σ is small. About 68% of the area under the curve occurs within $\pm$ one σ from the mean. Going $\pm 2\sigma$ from the mean will include better than 95% of the area under the curve. [By way of example, if a large number of measures of χ were made, 68% of these measures would fall within $\pm 1\sigma$ of the mean value, $\bar{x}$, and >95% would fall within $\pm 2\sigma$.] The use of $\pm 1\sigma$ is thus said to provide a confidence level (C.L) of 68%, $\pm 1.64\ \sigma$ gives 90% C.L., and $\pm 1.96\ \sigma$ results in a 95% confidence level. Clearly, specifying the precision limits of nuclear counting results is a matter of knowing the standard deviation σ.



In the normal curve which approximates the Poisson distribution, the standard deviation may be shown to be approximately equal to the square root of n the number of occurrences. That is:

$$\sigma \doteq \sqrt{n}$$

Since it is of more interest to know the error in the count rate (i.e. c/m) we divide by the time for the count:

$$\sigma \doteq \frac{\sqrt{n}}{t}$$

but n, the total count, = N t, where N is the count rate, so:

$$\sigma = \frac{\sqrt{Nt}}{t} = \sqrt{\frac{Nt}{t^2}} = \sqrt{\frac{N}{t}}$$

$$\boxed{\sigma = \sqrt{\frac{N}{t}}}$$

for 68% Confidence Level (C.L.) $\pm 1\sigma$
for 95% Confidence Level (C.L.) $\pm 2\sigma$

EXAMPLE: Report a sample which counted 5,000 counts in 4 minutes

$n_s =$ 5,000 counts

$t_s =$ 4 minutes

$N_s =$ 1250 c/m

$$\sigma = \sqrt{\frac{N}{t}} = \sqrt{\frac{1250}{4}} = \sqrt{312} = \pm 17.5$$

for 95% confidence level, use 2σ

therefore, $2\,(\pm 17.5) = \pm 35$

Reported value: $\boxed{N_s = 1250 \pm 35 \text{ c/m}}$

Suppose however, that in counting there is a background rate, or count rate measured when no sample is in the counter. Obviously, this background should be subtracted to obtain the count rate for the sample alone. But how are the respective errors combined? The theory of propagation of error states that if there is the error E_{S+BG} in the count rate of the sample-plus-background and the error E_{BG} in the determination of the background count rate alone, then the error E_{S+BG} in the net sample count rate will be square root of the sum of the squares of the individual errors:

$$E_S \text{ Net} = \sqrt{E_S^2 + E_{BG}^2}$$

EXAMPLE:

Given

$N_{S+BG} = 100 \pm 2$ c/m at the 95% C.L.

$N_{BG} = 20 \pm 3$ c/m at the 95% C.L.

Then the net sample rate $N_S = N_{S+BG} - N_{BG} \pm \sqrt{E_{S+BG}^2 + E_{BG}^2}$

$$N = 100 - 20 \pm \sqrt{2^2 + 3^2}$$

$$N = 80 \pm \sqrt{13}$$

$$\boxed{N = 80 \pm 3.6 \text{ c/m}}$$

(Note that simple addition of the individual errors in this case would have given a combined error too large by one part in four).

When individual errors are computed and combined in the above manner, the individual errors must be computed to the same confidence level. If it is desired to compute a combined error in a single step, the following formula may be used to obtain σ the standard deviation:

$$\sigma = \sqrt{\frac{N_{S+BG}}{t_{S+BG}} + \frac{N_{BG}}{t_{BG}}}$$

There may be derived a formula giving the most efficient distribution of counting time between counting background and counting sample. It is:

$$\frac{t_{S+BG}}{t_{BG}} = \sqrt{\frac{N_{S+BC}}{N_{BG}}}$$

EXAMPLE:

Given

$N_{S+BG} = 400$ c/m

$N_{BC} = 25$ c/m

$$\text{then } \frac{t_{S+BG}}{t_{BG}} = \sqrt{\frac{N_{S+BG}}{N_{BG}}} = \sqrt{\frac{400}{25}} = \sqrt{16} = 4$$

$\frac{t_{S+BG}}{t_{BG}} = 4$ or, in words, the ratio of sample counting time to background counting time should be four to one. Thus if one hour was available for the count, 12 minutes would be spent counting background and 48 minutes spent counting the sample.

It should be pointed out that the above formula assumes a "one shot" counting operation, i.e., that for each sample a separate determination of sample count rate and background count rate will be made. In usual counting-room practice, a background count is made only every two or three sample counts, and in some instances, only two or three times per day. In this case, the formula is not applicable without modification.

Since two successive counts of the same sample are not expected to give exactly the same count rate due to the random nature of the decay process, the question often arises as to how widely they may disagree and yet be in adequate statistical agreement. One agreement test for two successive counts is:

$$\sqrt{\frac{N_1}{t_1} + \frac{N_2}{t_2}} \leq K$$

For counting with a rate meter, Faires gives % statistical fluctuation:

$$\pm\,\% = \frac{100}{\sqrt{2RCN}}$$

where RC is the electrical time constant of the rate meter circuit, and N is the count rate in counts per second.

IV. PRACTICAL AIDS TO COMPUTATION

For a direct numerical approach, using the formulae discussed, a slide rule is usually sufficiently accurate for squaring and taking square roots. Computation may be considerably simplified by the use of nomographs found in the reference by Jarrett, the most useful of which are reproduced on pages 128-131 of the Radiological Health Handbook.* It covers counting rates from 1 to 100,000 c/m and counting times from 1 to 1,000 minutes, a range more than adequate to encompass the usual counting procedures. With this nomograph, a straightedge is used to connect the observed count rate (c/m) with the duration of the count (minutes), and the error at the 90% or 95% C.L. is read off the intersection with the center line, on the left or right side respectively. In the example solved previously numerically, the nomograph method gives a 95% C.L. error of ± 34.7 c/m for a sample which produced a count rate of 1250 c/m for 4 minutes. For background subtraction, the error in the background determination would be found in the same way, using the nomograph, and the individual errors from sample and background would be confined by squaring, adding the squares together, and extracting the square root.

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* 1960 Edition

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GPO 887-785

Sources and Reduction of Background of the Gamma Spectrometer

I. INTRODUCTION

Regardless of the type of radiation detection system (proportional, Geiger-Mueller, or Scintillation), one of the prime factors in determining the lower limit of detectability is the background of the system. This background, which is simply the count rate of the instrument when a sample containing no radioactivity is in the detector, is derived from three main sources and may be classified as follows:

A. Internal--radioactivity, both natural and man-made, that is present in the components of the detector system.

B. External--radioactivity and radiations that are located in the vicinity of the detector system such as natural activity in the construction material of the room, man-made contamination from a radiochemical laboratory, and cosmic radiations.

C. Electronic Noise--electronic pulses which may originate in the photomultiplier tube, amplifying, and analyzing circuits which are not due to interaction of radiation in the crystal.

A gamma scintillation detector presents several unique background problems which are not present in alpha or beta counters, mainly:

1. The mass of the detector's sensitive volume is relatively large.

2. Associated components of the primary detection device must be located physically close to it.

3. The detector has a relatively high efficiency for gamma and cosmic radiations which are difficult to shield against.

4. Placement of shielding material around the detector will change the shape of the net gamma spectrum obtained.

This outline will discuss the sources of external background and design of shielding to reduce their effect, the sources of internal background and methods for eliminating them, and typical background spectra for environmental counting.

II. EXTERNAL BACKGROUND AND ITS REDUCTION

The primary sources of external background are 1) cosmic radiations, 2) radon gas and its daughter products in the atmosphere, and 3) local natural and man-made radioactivity.

A. Cosmic Radiation

The earth is continually being bombarded by mesons and high-energy nuclei, predominately protons and alpha particles, which are called the hard component of cosmic rays. Their collisions with the nuclei of the atmosphere produce a large number of lower energy secondary radiations such as electrons, positrons, photons, and low energy mesons. The hard rays which make up approximately 80 percent of the cosmic radiation at sea level are extremely penetrating as shown by their ability to pass through the earths atmosphere which is approximately equivalent to 39 inches of lead. The rays also have been detected at a distance of 1400 meters of water equivalent below the earths surface. Therefore, artificial massive shielding is not practical, but neither is it needed, in most cases. An average "hard" cosmic particle will dissipate approximately 4.3 Mev per cm of path in a NaI crystal, and most will travel further than this in a crystal. Therefore, they cause little increase in the background of the energy range 0-4 Mev which is of interest in environmental analysis.

The other 20 percent of cosmic radiation, the "soft" rays, is easily absorbed by 4 inches of lead. But the net result from shielding is little reduction of cosmic rays because the few hard rays which do interact with the shield will re-emit secondaries within the shield.

The only effective methods for substantial reduction of cosmic background is the use of anticoincidence guard circuits which are roughly 30% efficient, or the use of great depths of natural earth shielding. Two hundred-fifty feet of earth shielding is used at Argonne National Laboratory which reduces the mesonic component by a factor of 70.

B. Radon Gas and Daughter Products

Radon gas, the immediate daughter product of radium-226 is present in all ambient air at concentrations ranging from 20 to 2500 pc/m^3. While radon is not a gamma emitter, its daughter products are and will contribute to the background in proportion to their activity concentration in the air. Their activity concentration is not constant and will vary with temperature, humidity, amount of air movement, and outside weather condition. If precautions are not taken, this activity can cause considerable daily variation in background. The most desirable remedy is to use aged, filtered air for ventilation with continuous recirculation and filtration. An alternative method is to use a high turn-over rate of filtered air from outside to prevent the buildup of daughter products.

C. Local Radioactivity

All building materials, such as a counting room would be constructed of, contain significant amounts of natural gamma emitters, particularly radium, uranium, thorium, their daughter products, and potassium-40. This activity alone will typically give a 0-4 Mev background for an unshielded 4" × 4" crystal of 10,000 cpm.

Artificial activity such as stored samples, stored isotopes, and fallout will contribute to the background. These sources should be eliminated as much as possible. However, their contribution along with natural local activity can be conveniently removed by the use of massive shielding.

III. DESIGN OF MASSIVE SHIELDING

The use of massive shielding surrounding the gamma scintillation detector is the primary means of drastically reducing the background. Shields ranging in weight from 1000 pounds to 50 tons have been designed for counting environmental samples. A typical shield would be in the 4 to 5 ton range. The shape, material, weight, and construction may be quite variable as seen in Figure 1.

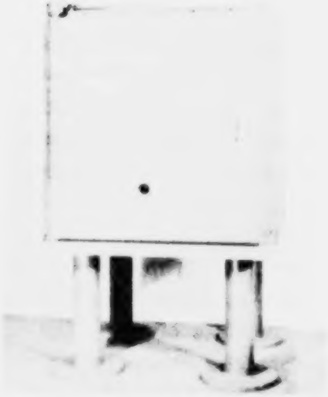
In the design of any shield for low-level detection the following factors must be considered:

1. Shielding properties
2. Radioactive content of shield material
3. Scattering by the shield material
4. Ease of construction
5. Economics

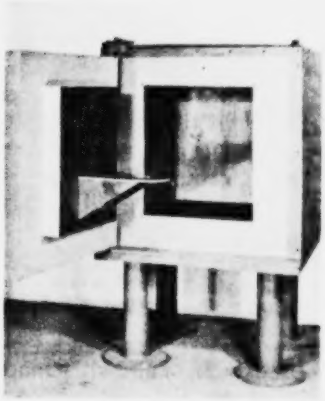
A. Shielding Properties

Gamma rays are absorbed exponentially according to the equation

$$I = I_0(b)e^{-\mu x} \qquad (1)$$



5-ton Steel Shield External View
Courtesy of Instrument &
Development Products Co.



5-ton Steel Shield Internal View
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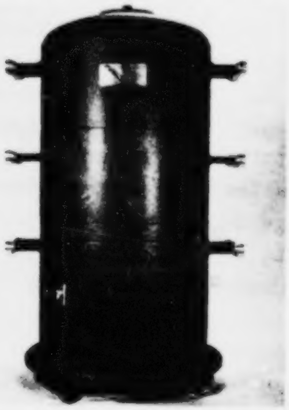
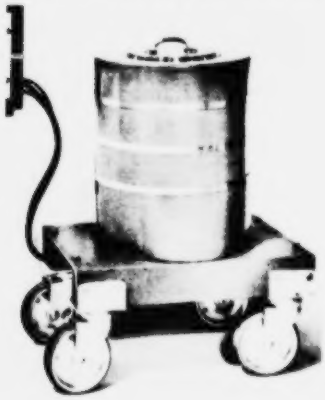
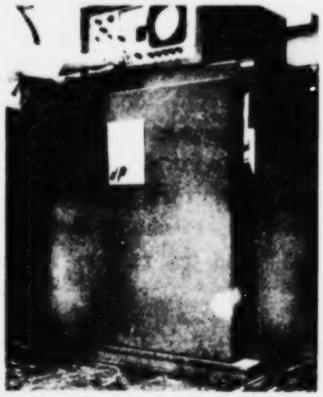


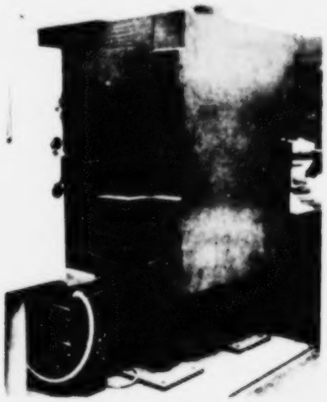
1000 Pound Lead Shield Module Construction
Courtesy of Instrument &
Development Products Co.



1000 Pound Lead Shield Mobile
Courtesy Victoreen Instrument Co.



4-ton Steel Shield
RHINO I



6-ton Steel Shield
HIPPO II

GAMMA DETECTOR SHIELDS
FIGURE 1

where:

I = Attenuated gamma flux

I_o = Initial gamma flux

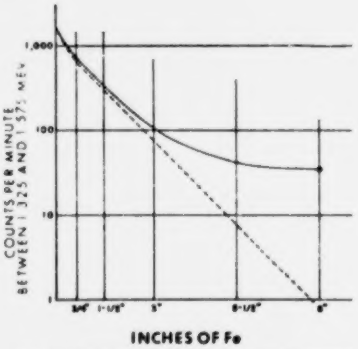
b = Build up factor

μ = Linear absorption coefficient

x = Thickness of shielding material

1. THICKNESS OF SHIELD

It would appear from equation 1 that by simply increasing the thickness (x) that any reduction of external background is possible. However, emperical curves, such as Figure 2, developed by measuring the background at various thicknesses of shielding and subtracting the background component from known sources, indicate that this is not possible.



BACKGROUND VERSUS THICKNESS OF SHIELDING MATERIAL

Figure 2

The dotted line shows the expected decrease in background, while the solid line indicates the observed. The asymptotic behavior suggests that practically all radiations due to laboratory surroundings have been substituted by the activity in the shield material itself. In the case depicted by Figure 2 the maximum shielding is obtained with approximately 6-7 inches of steel. In summary, the shield thickness will depend on the desired amount of external background reduction and will be limited by the activity concentration in the shield material.

2. SHIELD MATERIAL

The attenuation properties of any material is described by μ, the linear attenuation coefficiency in units of cm^{-1}, but perhaps two more meaningful concepts for comparative purposes are half-value thickness and equivalent thickness as they also involve the density of the material. Table 1 lists the properties of several common shielding materials.

It can be seen from the mass absorption coefficient, μ/ρ, that the shielding properties of material, on a weight basis, are approximately equal. However, when space is a prime consideration the higher density materials such as lead and mercury offer a definite advantage over a material such as standard concrete.

The theoretical lower limit of background will depend on the activity that is in the shield material itself. Therefore, the activity content of material is of prime importance in choosing a material.

B. Scattering Consideration

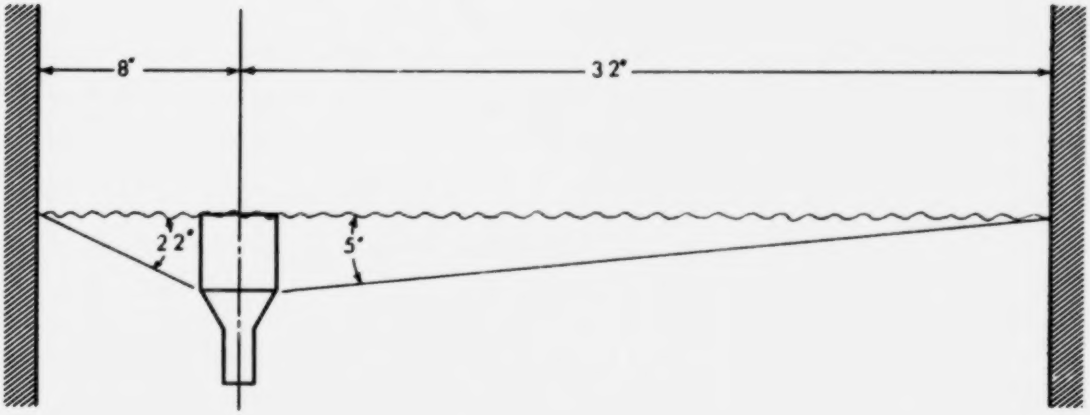
The most desirable counting system would be one which only produces a pulse height proportional to the energy of initial gamma photo emitted from the sample. In actual practice, photons and particles produced by scattering of the initial photon will result in pulse heights less than that of the photopeak. Two techniques, 1) shield configuration and 2) graded used to reduce the effects of scattering.

1. SHIELD CONFIGURATION

The majority of the scattered radiations from the shield is due to Compton interaction and is known as backscatter. While Compton

TABLE 1. SHIELDING PROPERTIES OF COMMON MATERIALS FOR 2.0 MEV GAMMA RAYS

	Density gm/cc	Equivalent Thickness (inches Fe)	HVL (inches)	μ (cm^{-1})	μ/ρ (cm^2/gm)
H_2O	1.00	6.50	5.45	0.050	0.050
Concrete	2.35	3.00	2.68	0.106	0.045
Steel	7.80	1.00	0.82	0.335	0.043
Lead	11.34	0.57	0.53	0.510	0.045
Mercury	13.54	0.48	0.44	0.624	0.046

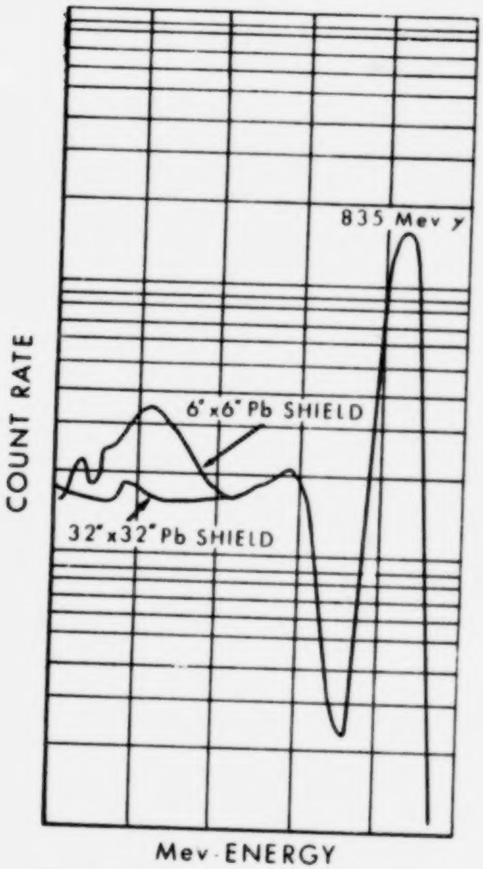


SOURCE OF 180° BACKSCATTER

Figure 3

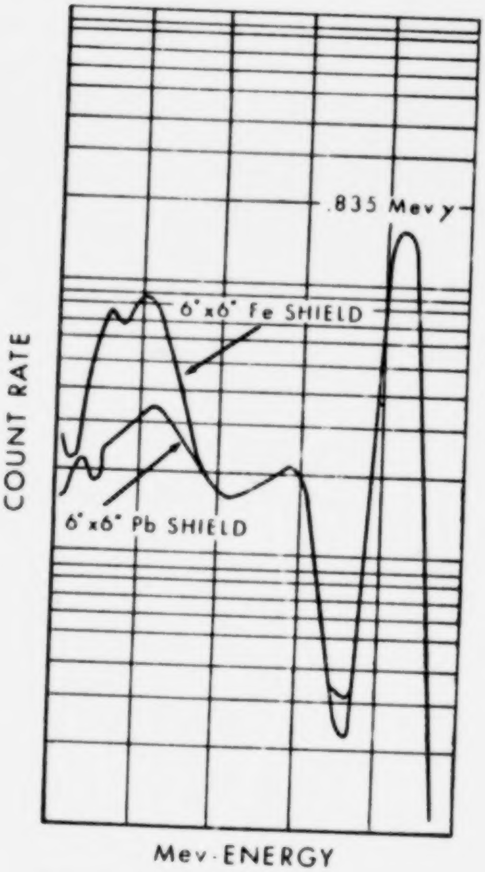
photons of all energies will be emitted from the shield, only those of a given energy range, corresponding to approximately 180° scatter, will interact with the crystal because of the particular geometrical arrangement of the source, shield and detector. Figures 3 and 4 illustrate this principle.

In Figure 3 it can be seen that a photon originating in the sample that interacts by Compton with the shield must be scattered back at approximately 180 degrees to be detected. The greater the distance the interaction takes place from the detector, the less is the scatter angle that will subtend the detector. Figure 4 shows the effect of greater distance on the shape of a Mn^{54} spectra; the backscatter component has to be drastically reduced. The conclusion is that the internal dimensions of the shield should be as large as cost and space will permit.



EFFECT OF SHIELD CONFIGURATION ON Mn^{54} SPECTRA

Figure 4



EFFECT OF SHIELD MATERIAL ON Mn^{54} SPECTRA

Figure 5

2. SHIELD MATERIAL AND LINERS

The cross section for Compton interaction decreases with increasing atomic number. Therefore, backscatter can also be reduced by use of high atomic number shielding materials. Figure 5 shows this decrease in a 6" lead shield compared to a 6" steel shield.

Photoelectric interactions give rise to low energy x-rays which are characteristic of the absorbing material. This x-ray production increases with atomic number and therefore is quite prominent in lead shields. Spurious radiation from this source is generally reduced by the use of "graded shielding". In lead shields, the shield is lined with one or more materials in descending order of atomic number. The materials are chosen to have a high cross section for the absorption of fluorescent radiation from the preceding one. A common liner is 1/16" of cadnium covered with 1/16" of copper.

Liners for steel shields are used for slightly different reasons. Unlike a lead shield, x-ray production in a steel shield is not significant, however, the Compton backscatter peak is. This can be reduced by use of a 1/8" lead liner to reduce the Compton photon production and to absorb those

Compton photons that are produced in the shield proper. Also the lead will slightly reduce the background from the activity in the shield material. However, the addition of the lead liner will introduce a lead x-ray peak which can in turn be reduced by adding a 1/16" stainless steel liner covering the lead liner.

C. Construction Considerations

Depending on the size, weight, permanency of location, and material; shields are usually constructed as hollow cylinders, or hollow cubes of solid or laminated walls.

1. CYLINDRICAL SHIELDS

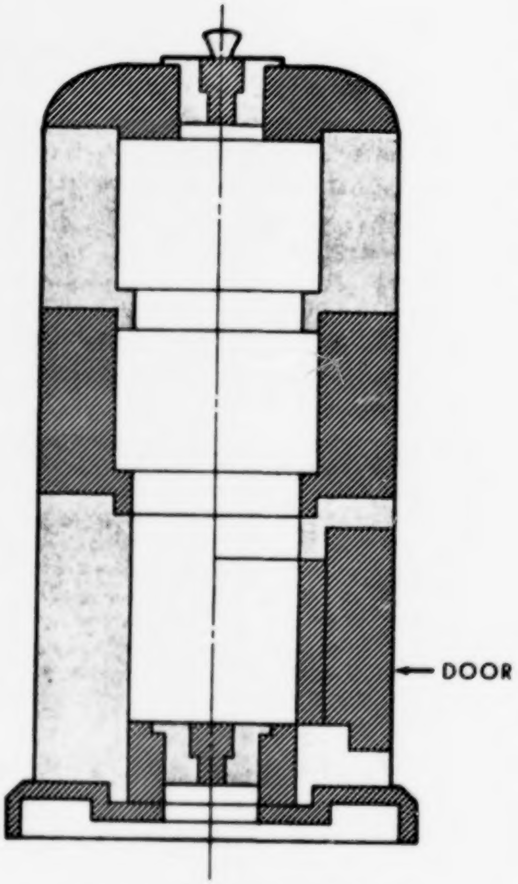
Smaller lead shields (1000 lbs. or less) commonly are cast as hollow cylinders in several modules such as shown in Figure 6.

The module construction with handles on each module greatly increase the mobility of the shield. Access to the inside can be obtained by either a side opening door as shown in Figure 1a or from the top as in Figure 1-b on page 3. Overlap construction on all joints and openings should be used to prevent radiation leaks.

2. CUBICAL SHIELDS

Larger shields, usually made of steel, are invariably constructed as hollow cubes. The use of laminated plates, 1/4" to 1/2" allows fabrication at the actual location of the counting facilities and eliminates the problem of handling very large weights. A 5 ton laminated shield is easily designed so that no one piece weighs more than 100 pounds. However, a frame is necessary to support the plates. A sheet metal dust cover is very desirable to reduce the possibility of contamination and to aid in decontamination. Overlap construction of the plates is very important when using relatively thin stock. The door which is usually one whole side of the shield and may weigh on the order of one ton presents special problems particularly in mounting. Heavy duty hinges such as a single 3/4" bar running the entire length of the door is usually necessary. Provisions for easily leveling the door to prevent drift is essential. A refrigerator type door latch is convenient to close and secure the door.

Solid shields have the advantage of less possibility of radiation leakage and are easier to fabricate. However, the transportation problem, particularly in tight quarters, is many times restrictive.

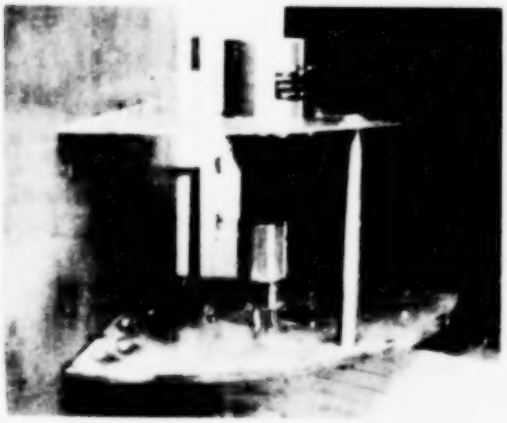


CYLINDRICAL LEAD SHIELD
OF MODULE CONSTRUCTION
Courtesy Instrument Development Products Co.

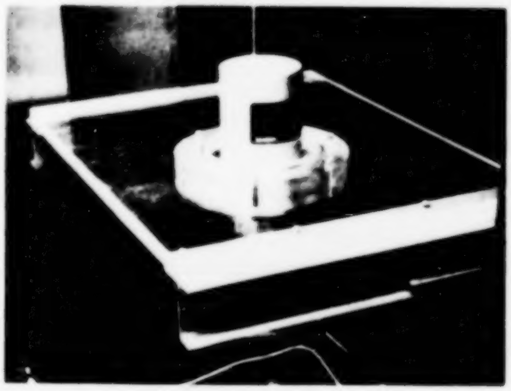
Figure 6

3. CRYSTAL MOUNTS

Two types of crystal mounting devices are used: 1) door mount and 2) platform mount. The door mount (see Figure 7) is very convenient for loading samples. However, severe physical shock may damage the crystal if the door is closed too abruptly. The use of a spring loaded door stop, will decrease this possibility. The platform mount (see Figure 8) which is simply a shelf on pullout slides eliminates the shock problem plus there is no chance of disturbing the sample when the door is closed.



DOOR MOUNT
Figure 7



PLATFORM MOUNT
Figure 8

IV. INTERNAL BACKGROUND AND ITS REDUCTION

While the background is most drastically reduced by the use of massive shielding, the background of even a well shielded detector can be reduced by a factor of two by awareness of internal sources of background and the proper selection of components to reduce this background. The factor of two can be the difference between the objectionable background level and a very tolerable level. The internal sources may be classified as follows:

1. Activity within the crystal proper.
2. Activity of the crystal container, optical reflector, and window.
3. Activity within the photomultiplier tube.
4. Activity within the shield material.

A. Crystal Activity

NaI crystals contain varying amounts of intrinsic potassium, some of which will be naturally occurring radioactive K^{40}. The similar chemical nature of K and Na makes it very difficult to obtain pure NaI without contamination from potassium.

Some NaI which has been used in the past for radiation scintillators has been found to contain as much as 55 ppm of elemental potassium. However, good quality crystals today may be obtained with potassium content of 1 ppm or less. Both the 1.33 Mev beta and the 1.46 Mev gamma will cause background pulses as the beta particles will not be shielded by the crystal can. Varying amounts of radium and thorium may be found in NaI also (NBS).

One(1) ppm/gm of elemental K in the crystal will result in approximately 0.013 cpm added to the background which would amount to 38 cpm/ppm K for a 4" × 4" (2940 gm) crystal. However, the beta count is distributed fairly evenly over an energy range of 0 - 1.5 Mev and a sharp photopeak is present only from the gamma ray.

B. Activity in the Crystal Container

The crystal container which is comprised of the can, reflector, and window may add significant counts to the background if precautions are not taken in selecting materials.

1. CRYSTAL CAN

The most commonly used canning materials are aluminum, copper and stainless steel. Aluminum presents the greatest possible source of contamination. Virgin aluminum contains small amounts of natural radium, thorium, and uranium but is not a major problem. However, most commercial aluminum is processed from scrap aluminum which invariably contains as much as 0.3 $\mu\mu$c of Ra/gm induced by man. This amount will add approximately 250 cpm to the background of a 4" × 4" crystal.

Stainless steel shows little contamination, while oxygen free copper is probably the cleanest of commercial metals.

2. REFLECTOR AND WINDOW

The standard reflector materials such as Al_2O_3 and MgO normally shows no detectable amounts of activity.

Windows are commonly made from glass, plastics, or quartz. Glass may contain significant quantities of natural K^{40} while plastics will contain natural or man-made C^{14}. Quartz, which has little activity, is used when low backgrounds are most critical.

C. Photomultipiler Tube

The photoassembly may account for as much as 20% of the background in a well shielded detector. The base of the phototube and the glass in the tube envelope are the major contributors.

The standard mica filled tube base can readily be eliminated by the manufacturer and the voltage divider wired directly to the leads from the tube.

Potassium and to a lesser extent radium, are normal constituents of the glass envelope. The practice of substituting vycor or fused quartz for part of the envelopment has greatly reduced the activity content. However, it appears that much more can and will be done in the future to reduce the level by substitution of low radioactivity materials.

D. The Massive Shield

The shield, which is used to reduce background from external sources, actually accounts for much of the total background. Natural Ra^{226} activity of steel is approximately 0.003 $\mu\mu$c/gram which for a 5 ton shield is 0.136 millicuries. Man-induced activities are fission products from fallout and Co^{60} from tracer pellets incorporated into fire bricks of the steel furnaces. For this reason great care must be taken in the selection and testing of steel to be used in shields.

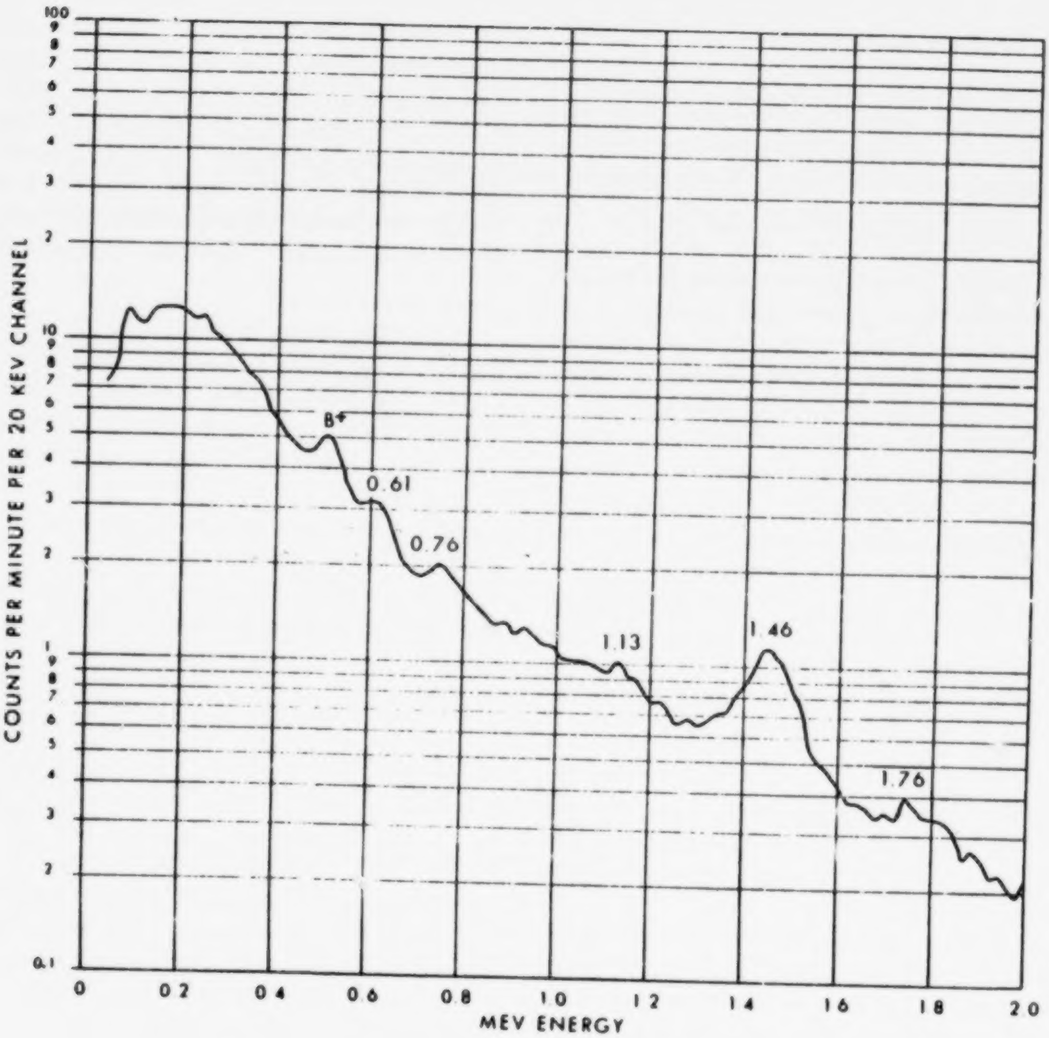
Recently, ruthenium-106 has been found in unproportionately high amounts. The exact source of this activity is not yet known.

V. BACKGROUND SPECTRA

Figure 9 is a composite background spectra for Detector System Hippo V located at the Robert A. Taft Sanitary Engineering Center, Radiological Health Research Activities, Low-Level Counting room. The detector system consists of a 4" × 4" NaI(Tl) Harshaw matched Window Line Assembly crystal mounted in a 24" × 24" × 36" - 6" steel shield with 1/8" lead, 0.020" cadmium, and 1/16" stainless steel liners. The shield is located three floors below ground level. A conserted effort has been made to reduce the background as much as is feasibly possible.

A. Background Count Rate

The 0.02 - 2 Mev background is 322 counts per minute with cesium channels (0.60 - 0.72 Mev) contributing 17.1 counts per minute.



BACKGROUND SPECTRUM FOR GAMMA SPECTROMETER HIPPO V
Figure 9

While a study of the percent contribution of various sources to this background has not been made, the literature indicates the major sources would be approximately as follows:

1. Shield material	30-40%
2. Cosmic rays	20-30%
3. Detector	15-20%

B. Prominent Features

There are three definite peaks in the spectra at 0.51 Mev, 0.61 Mev, and 1.46 Mev. The 0.51 Mev peak arises from the annihilation of cosmic positrons while the 0.61 Mev peak along with .72, 1.13, and 1.76 is due to radon daughter products. Potassium-40 accounts for the 1.46 Mev peak. The high background at low energies and the approximate power function decrease with increasing energy is typical.

VI. SUMMARY

The source of background are numerous and varied, but to effectively reduce the total background, they must all be considered. The primary ways of reducing background are the use of massive shielding and the selection of shield and detector components for low activity content.

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Statistical Evaluation of Gamma Spectra

I. INTRODUCTION

The basic concepts of nuclear counting statistics are applicable to gamma spectroscopy. Counting statistics can provide a baseline with which to judge the minimum error involved in quantitative gamma spectral analysis. This outline will point out some applications of nuclear counting statistics to gamma spectroscopy, and some limitations in its use.

Each channel of a multichannel analyzer can be thought of as a separate counting instrument. The data from each channel can then be treated in the same statistical manner as for an alpha or beta counting instrument. The statistical analysis of such data is routine but may not be directly applicable to multiple nuclide counting due to errors contributed by other nuclides.

II. SINGLE NUCLIDE COUNTING

The statistical evaluation of one channel of information can be done with the techniques already discussed. As one would expect, there are usually more than one channel's worth of useful information available for analysis. The use of several channels for a nuclide improves the counting statistics involved. One may consider the several channels as one large channel as for single-channel analyzer counting of a peak. There are several factors which dictate the number of channels chosen for analysis. These are the ratio of sample to background and the total number of counts.(1)

A high sample to background counting rate ratio will decrease the relative or percentage error.

EXAMPLE:

$$\%\ \text{Error at } 68\%\ \text{C. L.} = \frac{\sigma \times 100}{N_s} = \frac{\sqrt{\dfrac{N_{s+Bg}}{t_{s+Bg}} + \dfrac{N_{Bg}}{t_{Bg}}} \times 100}{N_s}$$

Case 1

$N_{s+Bg} = 30$
$N_{Bg} = 10$
$t_{s+Bg} = 10$
$t_{Bg} = 10$
$\frac{N_{s+Bg}}{N_{Bg}} = 3$

$$\%\ \text{Error at } 68\%\ \text{C. L.} = \frac{\sqrt{\frac{30}{10} + \frac{10}{10}} \times 100}{20}$$

$$= 10\%$$

Case 2

$N_{s+Bg} = 30$
$N_{Bg} = 5$
$t_{s+Bg} = 10$
$t_{Bg} = 10$
$\frac{N_{s+Bg}}{N_{Bg}} = 6$

$$\%\ \text{Error at } 68\%\ \text{C. L.} = \frac{\sqrt{\frac{30}{10} + \frac{5}{10}} \times 100}{25}$$

$$= 7.5\%$$

The peak channel itself has the highest sample to background counting rate ratio, however, the total number of counts is relatively small and better statistics arise from a greater total number of counts.

Case 1

$N_{s + Bg} = 30$

$N_{Bg} = 10$

$t_{s + Bg} = 10$

$t_{Bg} = 10$

$$\frac{N_{s + Bg}}{N_{Bg}} = 3$$

$$\% \text{ Error at } 68\% \text{ C. L.} = \frac{\sqrt{\frac{30}{10} + \frac{10}{10}} \times 100}{20}$$

$$= 10\%$$

Case 2

$N_{s + Bg} = 110$

$N_{Bg} = 50$

$t_{s + Bg} = 10$

$t_{Bg} = 10$

$$\frac{N_{s + Bg}}{N_{Bg}} = 2.2$$

$$\% \text{ Error at } 68\% \text{ C. L.} = \frac{\sqrt{\frac{110}{10} + \frac{50}{10}} \times 100}{60}$$

$$= 6.7\%$$

The decision between a narrow channel to get a better sample to background ratio and a wide channel to get larger numbers and better statistics can be resolved mathematically. Reber and Major$^{(1)}$ have analyzed the situation and have optimized the chosen channel width. They have assumed that the net peak has a normal distribution, and that the background is a linear function of energy. Figure 1 shows the assumptions involved.

It would be a laborious process to determine the standard deviation of the peak width by determining the deviation from the mean, in channels, for each individual count. Referring to Figure 1, if there are 20 counts in the peak channel, it means that in 20 determinations the analyzer found the energy of the photon to be that of the peak channel. The analyzer determined the energy of the photon to be that of each surrounding channel a lesser number of times. Altogether there are about 225 determinations (the peak area in counts). However, it has been determined that the full width half peak height of a normal curve (F.W.H.P.H.) is 2.36 times as wide as the standard deviation (σ), FWHPH = 2.36 σ. The FWHPH of a peak can be very easily measured from the plotted spectra so it can be used in place of σ.

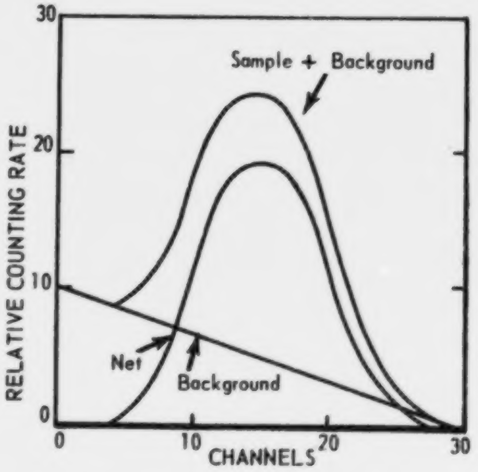

Figure 1

The optimum number of channels to be used for a given peak can be derived. A plot of such an optimization prepared from data of Reber and Major is given in Figure 2. The optimum number of channels to minimize statistical error is a function of the full width half peak height of the peak and the ratio (r) of sample plus background to background counting rates ($N_{s + Bg}/N_{Bg}$).

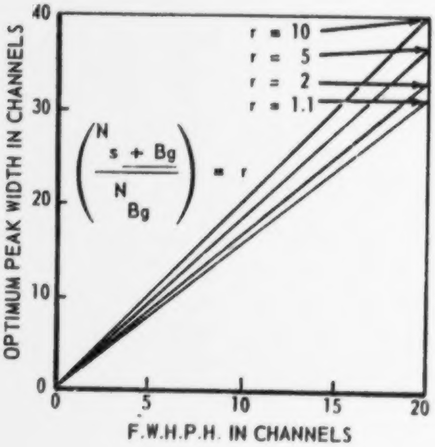

Figure 2

Figure 2 can be used to determine the statistically optimum number of channels to use for ratios of sample + background to background counting likely to be encountered. The full width half peak height is used as a measure of the spread of the photopeak.

III. PROPAGATION OF ERRORS

In elementary counting statistics, it is usually sufficient to be able to propagate errors where count rates are added or subtracted.

Many occasions arise when numbers and their associated error terms must be used in other operations, particularly multiplication and division. Therefore, it is also necessary to be able to propagate the errors in these situations.

A. Propagation of Errors in Multiplication

If it is necessary to multiply two terms, X and Y, both of which have an associated error term, E_x and E_y respectively, the error term E_z associated with the product is calculated as shown in equation 1.

$$E_z = X\,Y\sqrt{\frac{E_x^2}{X^2} + \frac{E_y^2}{Y^2}} \qquad (1)$$

A typical application is the prediction of a count rate when the sample disintegration rate and its associated error is known, and the counting efficiency which its associated error is known.

EXAMPLE:

Given

Sample disintegration rate = 40 ± 3.0 dpm at the 95% C. L.

Counting efficiency 0.60 ± .1 at the 95% C. L.

The predicted count rate and its error at the 95% C. L. is:

$$40 \times .6 + 40 \times .6\sqrt{\frac{3^2}{40^2} + \frac{.1^2}{.6^2}} = 24 \pm 4.4 \text{ cpm} \qquad 4$$

B. Propagation of Errors in Division

If it is necessary to divide one term, X, by another term, Y, and both terms have an associated error term, E_x and E_y respectively, the error term E_u associated with the quotient is calculated as shown in equation 2.

$$E_u = \frac{X}{Y}\sqrt{\frac{E_x^2}{X^2} + \frac{E_y^2}{Y^2}} \qquad (2)$$

A typical application is the determination of counting efficiency from a known standard disintegration rate with its associated error and the determined count rate and its associated error.

EXAMPLE:

Given

Standard disintegration rate = 90 ± 5.0 dpm at the 95% C. L.

Count rate determined = 40 ± 3.0 cpm at the 95% C. L.

The counter efficiency and its error at the 95% C. L. is:

$$\frac{40}{90} \pm \frac{40}{90}\sqrt{\frac{3^2}{(40)^2}+\frac{5^2}{(90)^2}} = 0.44 \pm 0.04$$

IV. ERRORS IN MULTIPLE NUCLIDE SPECTRA

The task of assigning an error term to each nuclide in a composite sample is considerably more complex than for a single nuclide sample. There is no simple way to evaluate the error contributions of the many factors involved in gamma spectroscopy.

Some of the factors which induce error in a spectroscopy analysis include instrumental gain shifts, instrumental zero shifts, statistical counting error of the peak nuclide itself, error in determining the contributions of other nuclides to that area, error in standards and calibration techniques, and errors in the mathematical model used for the analysis. The model is assumed to perfectly represent the physical situation, but this is not always true. For instance, unsuspected nuclides can invalidate the model. The only model where the errors are considered is in least squares quantitative analysis.

Some errors can be minimized by rigid quality control (such as instrumentation checks). Others may be partially defined or completely unknown.

The question "How much error should be attached to each nuclide determination in a mixture?" is an honest question that is yet to be completely answered. Computer programs are being written to estimate some minimum error term for the techniques such as matrices and least squares. As far as the general practitioner, the standby of standard samples or secondarily simulated samples for accuracy and replicate samples for precision estimates appears to be the best available means to provide an estimation of the overall error associated with an analysis.

V. MINIMUM DETECTABLE ACTIVITY

At levels of activity near background there is a question as to whether the indicated activity is real or merely normal variation in background. The concept of minimum detectable activity (MDA) has been developed to statistically answer this question.

The minimum detectable activity is defined as that amount of activity in counts per minute (cpm) which, in the same counting time, gives a count rate which is different from the background count by three (3) times the standard deviation of the background count.(2)

$$\text{M. D. A.} = 3\sqrt{\frac{N_{Bg}}{t_{Bg}}} \qquad (3)$$

where

t_{Bg} = $t_{s + Bg}$

N_{Bg} = count rate of background

t_{Bg} = counting time of background

$t_{s + Bg}$ = counting time of sample plus background

$N_{s + Bg}$ = counting rate of sample plus background

A second way of determining a lower limit for the detectable activity is to define the allowable relative counting error $\left(\frac{\text{Net count error}}{\text{Net count}}\right)$ at the required confidence level and back calculate to the activity necessary to achieve this error. The N_s is found by equation 4.

$$\text{Rel. Error} = k\sqrt{\frac{\frac{N_{s+Bg}}{t_{s+Bg}} + \frac{N_{Bg}}{t_{Bg}}}{N_{s+Bg} - N_{Bg}}} \qquad (4)$$

where

k = multiple of σ corresponding to the desired confidence level

In this approach, the required precision is defined and the minimum activity in cpm that will have this counting error is calculated.

VI. MINIMUM SENSITIVITY

The concept of minimum sensitivity is directly applicable to an analysis situation. Minimum sensitivity (M.S.) is defined as the activity concentration in pc/volume which is detectable under specific conditions of sample volume, background, counting time, chemical yield, and counting efficiency.

$$\text{M.S.} = \frac{\text{M.D.A.}}{(\text{eff.})(\text{chem. yield})(2.22\ \text{dpm/pc})(\text{size})} \qquad (5)$$

It may also be useful at times to determine the lowest activity concentration that can be determined with a given precision. A determination of this type is given in example 1.

EXAMPLE 1:

Required: The lowest activity concentration of Fe^{59} in a sample where the desired precision is 25% at 95% C. L.

Given: Sample volume = 250 ml

Background = 8.5 cpm

Background Counting Time = 100 minutes

Sample Counting Time = 100 minutes

Efficiency (1.097 peak) = 0.00795

Chemical Yield = 75%

Solution: The desired error is defined by

$$\frac{\text{Error in cpm}}{N_s\ \text{in cpm}}(100) = 25\%$$

Use equation 4 to determine the activity concentration.

$$2\sqrt{\frac{\frac{N_s + 8.5}{100} + \frac{8.5}{100}}{N_s + 8.5 - 8.5}} = 0.25$$

In a simplified form the above equation is:

$$2\sqrt{\frac{\frac{N_s + 8.5 + 8.5}{100}}{N_s + 8.5 - 8.5}} = 0.25$$

Solving this expression yields

$$N_s = 3.64\ \text{cpm}$$

The lowest concentration activity is calculated by converting N_s in the same manner as MDA in equation 5.

$$\text{Activity Concentrations} = \frac{3.64}{(.00795)(.75)(2.22)(.25)}$$

$$= 1{,}100\ \text{pc/l}$$

VII. SUMMARY

Basic nuclear counting statistics can be applied to gamma spectroscopy. For a single nuclide the number of channels chosen determines the relative statistical error. In multinuclide mixtures, the apportionment of error among the many contributors is not feasible for the ordinary practitioner. Application of statistical techniques to standard and replicate samples probably provides the best available estimate of error for each nuclide in the sample. The application of the minimum detectable activity (MDA) concept statistically determines whether a near background activity is real or probably due to fluctuations in background.

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Quality Control of Analysis by Gamma Spectroscopy

I. INTRODUCTION

In any type of analytical work the analyst should continually ask such questions as "how good am I doing," or "am I getting the right results?" It is not enough that these questions should be continually asked, but they should also be continually answered. The use of a systematic approach to answer these questions is known as a quality control procedure. It will be the purpose of this discussion to point out several of these procedures that can be used in analysis by gamma spectroscopy.

It should be emphasized at the outset that quality control should not be inserted in an analysis program as an afterthought. It requires additional analysis, data calculation, and administration; all of which should be considered in setting up personnel schedules and workloads. This should not be considered as a pure overhead cost, as it may well result in the savings of large sums of money otherwise lost due to continuing errors not found until too late.

II. DISCUSSION OF ACCURACY

As stated above, the objective of quality control is to give reasonable assurance that correct analytical results are being obtained. Probably a better nomenclature would be that we are interested in the accuracy of our results. The accuracy of an analysis depends on two parameters: precision and bias.

A. Precision

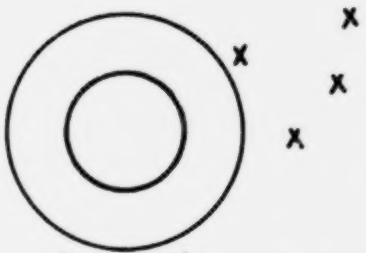
Precision means the closeness of approach of a number of similar measurements to a common value. For our purposes, precision is measured in terms of deviations of the individual measurements of a series from the average value of that series. Precision is not only very necessary to obtain accurate results but is also an excellent measure of the worth of the analysis method being used. If a reasonable precision cannot be obtained by the method, accuracy can never be obtained.

B. Bias

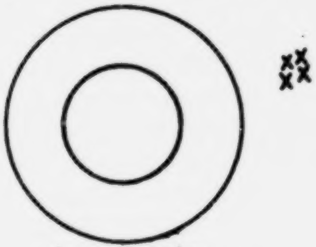
Although a high degree of precision may be obtained, this does not assure accuracy. All of the replicate results of a determination may closely agree with each other and still be far removed from the correct result. This may be due to an inherent feature of the analysis method or a systematic error in carrying out the procedure.

One classical way of illustrating both precision and bias is to use target diagrams, see Figure 1. This figure shows the different combinations of precision and bias that may result with four replications. An X represents the analytical result of one determination and the bulls eye of the target represents the correct result.

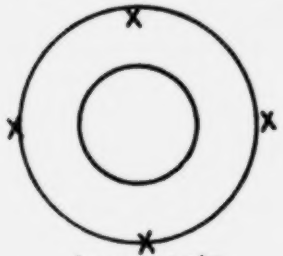
A quality control program is intended to allow a check on both precision and bias so that difficulties may be quickly detected and



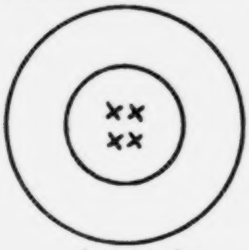
Inaccurate
Imprecise and Biased



Inaccurate
Precise, but Biased



Inaccurate
Unbiased, but Imprecise



Accurate
Precise and Unbiased

Precision bias and accuracy
Figure 1

the causative factor corrected. This should give continuously accurate results.

III. DISCUSSION OF ERRORS

Departures from accuracy may result from many different occurrences. We will discuss these errors in two categories: "Determinate errors," and "Indeterminate errors."

A. Determinate Errors

Determinate errors are those which can be given an assignable cause. These may in turn be sub-categorized as follows:

1. PERSONAL ERRORS

These are errors which are attributable to individual mistakes made by the analyst. This may result in imprecise and/or biased errors.

2. INSTRUMENTAL ERRORS

Any change due to instrumental components which alters detection efficiency or response will fall in the category of instrumental errors and may result in an extreme bias.

3. ERRORS IN METHOD

The use of an incorrect calibration standard, chemical preparation technique, or data treatment method would all fall in this category of error.

It is the purpose of quality control to detect all determinate errors. On the basis of this detection the attributing cause can then be sought.

B. Indeterminate Errors

Even when all determinate errors are eliminated every replicate analysis will not give the exact same value. This is especially true in the nuclear field as the results usually are derived from some radiation counting technique with its inherent statistical fluctuation. The other deviations in this category may be due to slight variations in judgment such as reading of measuring devices.

Indeterminate errors, or perhaps a better nomenclature would be indeterminate variations in results, are something that we have to live with. Fortunately these indeterminate variations conform to the laws of statistical distributions so that they can be handled mathematically with results stated as a narrow range of possible true values, with certain confidence limits.

One of the major problems that does arise due to indeterminate errors is the increased difficulty of detecting determinate errors (causal errors) in the presence of continually fluctuating values. It is to this latter end that much of our quality control work in gamma spectroscopy is directed.

IV. QUALITY CONTROL APPLIED TO THE THE GAMMA SPECTROMETER

In radionuclide analysis by gamma spectroscopy the entire validity of the analysis depends on the satisfactory operation of the gamma spectrometry instrumentation. The first level of quality control then is that applied to the instrument itself.

A. Reference Standards

It would be entirely too laborious to redetermine instrument detection efficiencies every time that an analysis is run. Not only would this be much too time consuming but it would require an inordinate number of standards to be kept on hand which would be most difficult, especially for short half-life material.

The procedure that is usually adopted is to make a daily count of a reference standard for which an accurate count was taken when the instrument efficiency was determined. This then becomes a daily control check that the instrument's efficiency has not changed. The most predominately used reference standard for gamma spectrometer systems is a source of cesium-137. In multi-channel systems the control count is usually made by summing up the .662 Mev peak channels, while in single channel systems the window is usually set to cover this peak or an integral count is made. It is good practice to use two reference sources on each instrument so that it can be immediately determined that deviations that occur are not due to leaking, contamination, or other problems with the reference standard. The two major requirements of reference standards are that they contain sufficient activity to give good statistics in a reasonably short time and that the isotopic content is pure enough that the decay can be accurately determined. It would also be recommended that one of the reference standards be a point source, or nearly so, and the other would be an extended source that would cover the crystal face. Such a difference in geometry of check sources would be of great assistance in detecting a cracked crystal or some non-uniform response that might occur.

The use of such a reference standard as has been discussed is not completely foolproof. Instrument errors which might be restricted to precise energy ranges, such as errors due to malfunction of certain storage locations or readout, would not show up unless the check source summed counts happened to fall in this energy or pulse height range. A check on the complete energy span can be made by the methods described under background quality control.

B. Use of Control Charts

As there will be a certain indeterminate error in our reference standard counts, primarily due to counting statistics, it is necessary to be able to account for these and still be able to detect deviations due to some causal effect. This is usually done by recording the daily reference standard count on a control chart.

The control chart usually consists of a graph with time on the abscissa and count rate on the ordinate. Heavy lines are drawn at count rates representing the best estimate of the true count rate (corrected for decay if necessary) and plus and minus 1, 2, and 3 times the standard deviation of this "true count rate" value. The standard deviation is calculated from the "true count rate" value for the time interval in which the daily counts will be made. The "true count rate" is probably best determined by taking the average of about 20 countings that have good individual statistics and are normally distributed about the average.

Figure 2 shows the general set up of a control chart that might be used.

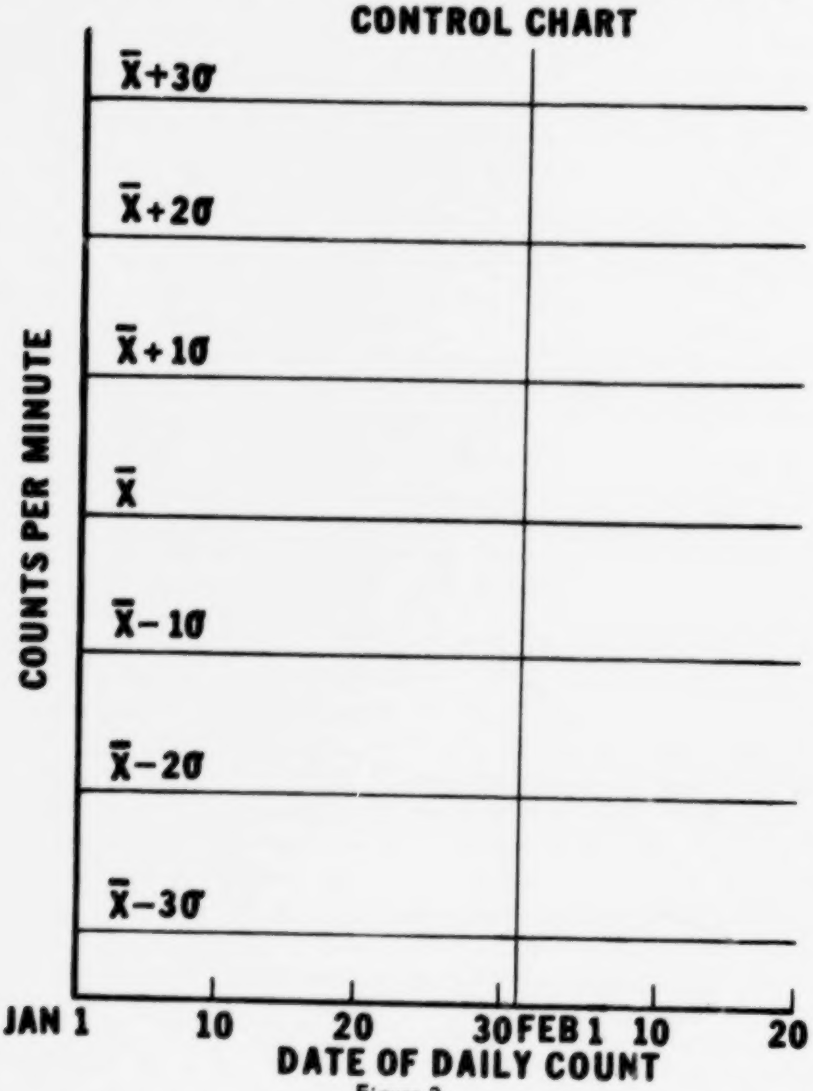


Figure 2

Such a control chart as this can be a valuable guide as to the stability of the operation of the instrument. The number of determinations falling within the different standard deviation limits can be periodically checked to see if they can conform with the associated theoretical probability, and divergent trends can be readily detected. Any time that an inordinate number of reference standard counts fall outside the 2 standard deviation limits an assignable cause should be sought and the situation corrected.

C. Chi Squared Test

Although the use of control charts is a convenient method for daily checking with reference standards it does not always provide the answers in as quantitative a manner as is desirable. It may be desirable to periodically check the accumulated daily reference counts to more rigorously determine if all the data belongs to the same statistical normal distribution. One method of answering this question and being able to attach a probability to the answer is by the application of the Chi Square (χ^2) test.

The Chi Square test actually tests the distribution of differences of individual values from the mean. The numerical value of Chi Square (χ^2) for a set of data is calculated as shown in equation 1.

$$\chi^2 = \frac{\sum_{i=1}^{i=m} (\bar{X} - X_i)^2}{\bar{X}} \qquad (1)$$

This shows that the numerical value of Chi Square is the sum of the squares of the differences between the mean of true value, $\bar{X}$, and the individual values, X_i, with the summation divided by the true value, $\bar{X}$. For each value of Chi Square with the same number of replications, m, there is a specific probability. These probabilities can be obtained from Table 1.

Both a very low probability and a very high probability may be indicative of nonconformance to a normal distribution. Quite often the limits of acceptance are set at both extremes. For instance, the upper limit might be set at .95 and the lower limit at 0.05.

EXAMPLE:

In order to check the reproducibility of a counting system, 10 one minute counts are made. Are these 10 counts all part of the same normal distribution? 1) The values of the individual one minute counts, X_i, are shown in the first column of the table below:

Number of counts in 1 min. interval (X_i)	$\bar{X} - X_i$	$(\bar{X} - X_i)^2$
29	3.8	14.44
36	10.8	116.64
19	6.2	38.44
26	0.8	.64
24	1.2	1.44
14	11.2	125.44
21	4.2	17.64
32	6.8	46.24
24	1.2	1.44
27	1.8	3.24
SUM 252		SUM 358.20

$$\text{The mean } \bar{X} = \frac{\sum_{1}^{10} X_i}{M} = \frac{252}{10} = 25.2$$

$$\chi^2 = \frac{\Sigma (\bar{X} - X_i)^2}{\bar{X}} = \frac{365.60}{25.2} = 14.51$$

From Table 1 it is seen that this series of determinations has a probability of greater than 0.10 of being a normal distribution.

Table 2. Table of CHI-Squared

Number of Determinations	Probability						
	0.99	0.95	0.90	0.50	0.10	0.05	0.01
	Values of Chi-Squared						
2	0.020	0.103	0.211	1.386	4.605	5.991	9.210
3	0.115	0.352	0.584	2.366	6.251	7.815	11.345
4	0.297	0.711	1.064	3.357	7.779	9.488	13.277
5	0.554	1.145	1.610	4.351	9.236	11.070	15.086
6	0.872	1.635	2.204	5.348	10.645	12.592	16.812
7	1.239	2.167	2.833	6.346	12.017	14.067	18.475
8	1.646	2.733	3.490	7.344	13.362	15.507	20.090
9	2.088	3.325	4.168	8.343	14.684	16.919	21.666
10	2.558	3.940	4.865	9.342	15.987	18.307	23.209
11	3.053	4.575	5.578	10.341	17.275	19.675	24.725
12	3.571	5.226	6.304	11.340	18.549	21.026	26.217
13	4.107	5.892	7.042	12.340	19.812	22.362	27.688
14	4.660	6.571	7.790	13.339	21.064	23.685	29.141
15	5.229	7.261	8.547	14.339	22.307	24.996	30.578
16	5.812	7.962	9.312	15.338	23.542	26.296	32.000
17	6.408	8.672	10.085	16.338	24.769	27.587	33.409
18	7.015	9.390	10.865	17.338	25.989	28.869	34.805
19	7.633	10.117	11.651	18.338	27.204	30.144	36.191
20	8.260	10.851	12.443	19.337	28.412	31.410	37.566
21	8.897	11.591	13.240	20.337	29.615	32.671	38.932
22	9.542	12.338	14.041	21.337	30.813	33.924	40.289
23	10.196	13.091	14.848	22.337	32.007	35.172	41.638
24	10.856	13.848	15.659	23.337	33.196	36.415	42.980
25	11.524	14.611	16.473	24.337	34.382	37.382	44.314
26	12.198	15.379	17.292	25.336	35.563	38.885	45.642
27	12.879	16.151	18.114	26.336	36.741	40.113	46.963
28	13.565	16.928	18.939	27.336	37.916	41.337	48.278
29	14.256	17.708	19.768	28.336	39.087	42.557	49.588

Although the Chi Squared test may not be adopted as a routine procedure it can be a most useful additional check.

D. Resolution Quality Control

The methods of instrument quality control that have been discussed so far are mainly a check on instrument detection efficiency and to some extent stability. Many of our spectrum analysis methods (e.g., spectrum stripping) are very dependent on the reproducibility of the spectrum shapes, especially the shape of the photopeak. So this is another parameter that requires quality control checks.

The usual check that is made is a check of the instrument resolution for some designated photopeak, with the 0.662 Mev Cs-137 photopeak being the one that is most often used. It may be sufficient to make a daily or weekly determination of resolution and compare with past values. The tolerances that can be accepted on changes in resolution will depend to some extent on the analysis method to be applied. One laboratory has set up a criteria of accepting a 0.2% change in percent resolution with changes of 0.5% considered to be well out of control as this would cause a considerable error in quantitative analysis.

One of the refinements that has been applied to resolution quality control is to plot the photopeak distribution on probability paper. In this case it is necessary to convert the count rate data of the photopeak channels into cumulative integral counts starting with the lowest energy channel of the photopeak. The cumulative counts are then converted to percentages of the total photopeak counts and these percentages are plotted vs the pulse height or channel number on probability paper. See Example 2. This should come out to be a straight line plot which is representative of the distribution of the counts in the channels considered. In the case illustrated in example 2 the criteria for picking the considered channels was that they include a greater portion of the peak than that of 1/2 the peak height and that they do not include any of the area where substantial deviations from a normal distribution of count rate with channel occur. (The method assumes a gaussian distribution for the photopeak). A probability plot such as this can provide a convenient method of comparing photopeak shapes from time to time. The line should be reproducible each time regardless of the cesium standard count rate or whether total counts or counts per minute is plotted as it is based on the percentage of the total. Any change in the slope of the line will be due to an actual change in resolution or a gain shift.

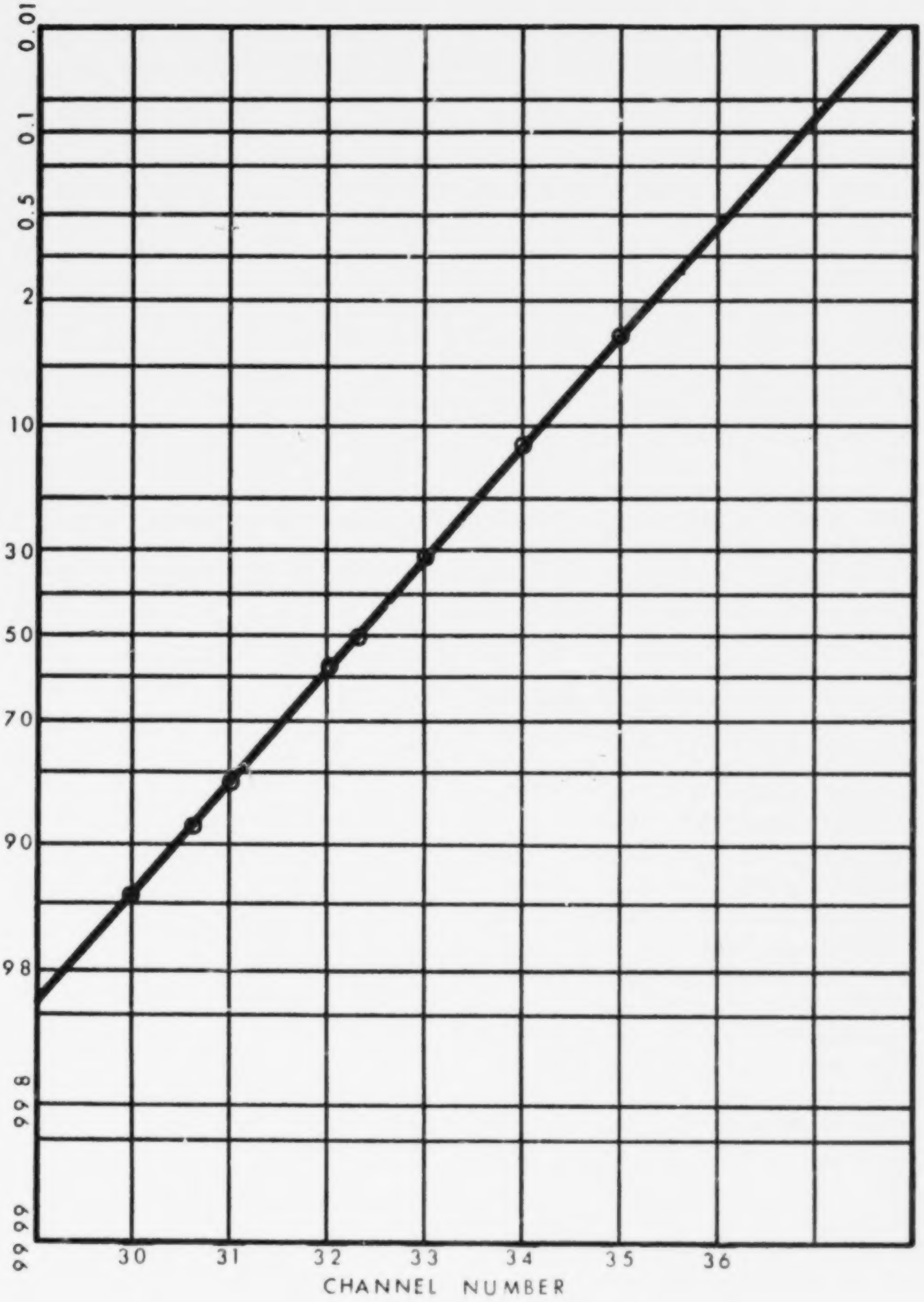
EXAMPLE 2: The count rate data shown below was obtained for the selected photopeak channels of Cesium-137. Make a plot of this photopeak on probability paper.

Channel No.	CPM	Integral Counts	% of Total
30	14.56	14.56	5.6
31	34.35	48.91	18.7
32	63.05	111.96	42.9
33	67.05	179.01	68.6
34	50.14	229.15	87.8
35	23.51	252.66	96.8
36	8.32	260.98	100

Figure 3 shows a plot of the % of total vs channel number. Note the 100% point (channel 36 in this case) is not plotted when doing this type of analysis.

V. BACKGROUND QUALITY CONTROL

It is not sufficient to check only the instrument's response by use of reference standards as has been described above. Another major parameter related to the instrument, but not wholly dependent on it, is the



Probability plot of Cs-137 photopeak
Figure 3

background level that is obtained. Although the quality control of background can be handled very similarly to the methods used with reference standards, some special precautions should be taken.

A. Control Charts for Background

In general the control chart technique mentioned in connection with reference sources can be applied to background. This might be particularly suitable with single channel systems where repetitive integral counting or counting over some prescribed energy range is used. The charts may also be used with multi-channel systems where several channels are summed.

There is one major difference between these control charts and ones for reference standards. Where in the case of reference standards the standard deviation can be determined purely on the basis of counting statistics, i.e., the square root of the true count, this technique is not usually sufficient for the background in gamma spectrometer systems. The background does not only fluctuate due to counting statistics but superimposed on this is another fluctuation which is due to the fluctuation of actual physical background itself, i.e., primarily radon in gamma detection systems. Although this fluctuating background can be greatly reduced it still must be considered.

The mean and standard deviations (σ) to be used on a background control chart should be calculated by normal statistical techniques, using equations 2 and 3 respectively, from several months of data which include the most predominate seasonal variations to be encountered.

$$\overline{X} = \frac{\sum_{i}^{n} X_i}{n} \qquad (2)$$

$$\sigma = \frac{\sum_{i}^{n} (X_i - \overline{X})^2}{n} \qquad (3)$$

The background determinations are usually made overnight to assure adequate counting statistics. However, there should be comparison tests made between the day and night background to assure that the differences are not significant enough to cause bias in the final results. The gamma spectrometer background control chart will not show a random spread of values as will the control chart for a reference standard. With changing atmospheric conditions it may even resemble a sine wave or some other oscillating function. Although these considerations certainly reduce the value of a background control chart the detection of some contamination such as K^{40} would still probably show up quite obviously.

B. Other Background Control Techniques

The use of control charts for background does not fulfill all the needs and other checks must be made on background levels. The control chart will only indicate changes in the gross spectrum or segments of the spectrum for which the counts are summed. This would enable the detection of contamination from radionuclides with gamma energies higher than the energy span plotted but would give no information if the contaminant had a lower energy. It is also necessary to keep an eye on the consistency of the background counts as they are distributed with energy. One way to make this comparison is to compare the graphical background by overlay on a light table or some similar arrangement.

A standard background comparison curve can be constructed by plotting the all time high and low backgrounds to form a band within which the daily background can be expected to fall. Changes in the background energy distribution can be detected quite easily in this manner and may lead to early detection of shield contamination or other difficulties.

As the fluctuating radon level is such an important influence on the background changes, one technique that has been suggested is to install an external radon counter in the counting room so that these fluctuations can be recorded and taken into consideration when they change appreciably.

It should be mentioned that the Chi-Squared test may also be applied to background counts in the same way that it was applied to reference standards. This will often give a useful indication of just how random the background fluctuations are and help in determining the proper control method.

VI. CONTROL CHECKS FOR SAMPLE CONTAINERS

The usual counting facility will use a number of different types of sample containers, some of which will be purchased in large lots. There is always the problem in cases of this type of assuring that the containers are identical in the necessary parameters. At least two types of uniformity checks should be made on containers, one for background and one for geometry. Control charts and the Chi-Squared test can be adapted to both of these needs, where in this case each container counted would represent a point on the abcissa rather than a daily count.

In checking container background, a few containers from each lot should be selected at random and counted. The variation as shown on the control chart should be no greater than that expected for natural background fluctuation. It is also good procedure to check the shape of the spectrum for the sample containers. This can be done by the same techniques applied to background in general.

The geometry of containers can be checked by counting solutions of known activity and again plotting the results on a control chart with acceptable results falling within expected counting statistics.

The matter of how many containers should be tested from each lot will depend on the size of the lot and the degree of confidence desired. The ultimate, of course, would be to test every container, but this is not feasible so some compromise must be accepted.

For some types of non-disposable containers it may be necessary to keep a separate background record on each one. This might be the case when a Marinelli beaker is used for counting rather high activity standard solutions. In some cases certain beakers are set aside and only used to count one type of standard solution. This is especially necessary where plating out of radioactive material is a problem.

These quality control checks for sample containers should not be confined to an initial check when they are received but should be repeated periodically for containers which are used more than once.

VII. QUALITY CONTROL OF TOTAL ANALYSIS

Even though a number of quality control checks can be initiated in relation to specific steps it is well also to keep a check on the overall results.

A. Precision Quality Control

Once an analysis method has been thoroughly checked out and found to give accurate results for all those using the method a good deal of control can be exercised by checking precision of results. This can best be done by submitting replicate (at least duplicate) samples in all cases. The person who is counting the samples and those analyzing the spectrums should not be aware of which ones are duplicates. It is often advisable to split up the duplicates and have them submitted on different days. This will also give an indication of day to day changes. Such a system will quite often show up any difficulties that occur. Those administering the quality control program should be aware of what precision the analysis method is capable and what variation they will accept. It may be found that after keeping a check on duplicate samples for a period of time that these acceptable limits of variation may be narrowed due to actual performance well within them. In some cases the allowable variation between replicates will depend on the activity of the sample. With higher activity the absolute variation may be higher but still well within method capabilities. For instance, the limits of acceptability might be set as ± 5% or ± 5 picocuries per liter, whichever is greater.

B. Accuracy Quality Control

As previously discussed, the maintenance of precision is not conclusive of accuracy. It may be that some change in method or other error has occurred which is resulting in the results being biased. It is well to check the overall accuracy of the analysis at periodic intervals by submitting check samples of known radionuclide content.

In many cases it may be quite difficult to make up a simulated environmental sample of known radionuclide content. One procedure that can be used is to have a large environmental sample on which sufficient cross checks have been made that the actual content is known. Aliquots from this large sample can then be used as check samples for subsequent analysis.

The number of check samples that are analyzed will depend on overall workload and degree of assurance desired. It is not unusual for some laboratories to have 15% of their samples as check samples. Again the individuals doing the counting and analysis of data should not be aware that they are processing anything other than an ordinary routine sample.

VIII. SUMMARY

Quality control is a necessary part of any analysis process and gamma spectroscopy is no exception. Control checks should be applied to instrumentation variables as well as other points in the analysis schemes. The final results should be checked by replicate and check samples to determine overall precision and accuracy. Only through a comprehensive program can one be assured of the quality of analytical work done.

Selection of Gamma Standards

I. INTRODUCTION

The selection of a radionuclide standard for determining counting efficiencies is one of the most important steps in any radioactivity determination. Unless proper care is exercised in selecting and using the standards, the final results obtained may be of little or no value.

This discussion presents information relating to the procurement, quality and properties of gamma counting standards. Particular emphasis is placed on the preparation of gamma counting standards and the interpretation of the instrument efficiency which is obtained from the use of gamma standards.

II. PROCUREMENT OF STANDARDS

Artificial, naturally occurring radionuclides, and stable nuclides may be obtained from various governmental and private organizations whose publications(1, 3) should be consulted for lists of materials and services furnished.

Artificial radionuclides are produced by bombardment of suitable target materials either with neutrons in a nuclear reactor or with charged particles in a cyclotron or other type of accelerator. Certain radionuclides may be obtained as fission products from nuclear reactors. Naturally occurring radionuclides are made available through chemical separation and purification processes.

III. QUALITY OF STANDARDS

A. Accuracy

The major errors affecting the accuracy of a standard are due to unstable chemical and physical properties, dilutions, source preparations, counting statistics, counting losses and corrections, decay scheme assumptions, and the initial calibration and subsequent consistency of the standardization instrument. These factors contribute to the error specification reported by the supplier. They are important in evaluating the accuracy of an instrument calibration and comparing similar standards in duplicate calibrations. The error associated with the accuracy of a standard is stated in numerous ways. Table 1 presents error statements reported by three national and two foreign suppliers.

Only foreign supplier #2 distinguishes between precision and systematic errors, presents values for the types of errors considered, and indicates the confidence levels at which the values are derived. The other four suppliers do not furnish this information and thus the user is unable to determine the full significance of the error term. Difficulties have arisen in the comparison of calibrations of radionuclides from one supplier to another because of the differences in certificate formats.(4)

B. Purity

As in the case of beta standards, small quantities of contamination may be accepted in gamma standards under certain conditions. These conditions usually relate to the qualitative and quantitative data available concerning the contamination.

Impurities present in gamma standards may cause discrepancies in counting efficiency values along with distortions in spectral shapes. For example, since ruthenium-106 and ruthenium-103 possess similar principal photopeaks, the presence of ruthenium-106 in a fission-produced ruthenium-103 standard interferes with an accurate calibration and spectrum analysis effect. Other activities that may be present in commonly used gamma standards are illustrated in Table 2.

Table 1[4]: Typical error statements of suppliers of radioactivity standards

---

U.S. SUPPLIER #1:

"Specific activity is.......dps/ml ± 3%."

U.S. SUPPLIER #2:

"The estimated overall accuracy of the standardization is ± 2%."

U.S. SUPPLIER #3:

"The accuracy of the standardization is estimated to be ± 2%."

FOREIGN SUPPLIER #1:

"Overall error ± 1%."

FOREIGN SUPPLIER #2:

"Accuracy:

The 'EFFECTIVE STANDARD DEVIATION' (Defined as 1/6 of the overall range between the confidence limits stated below) is less than two%.

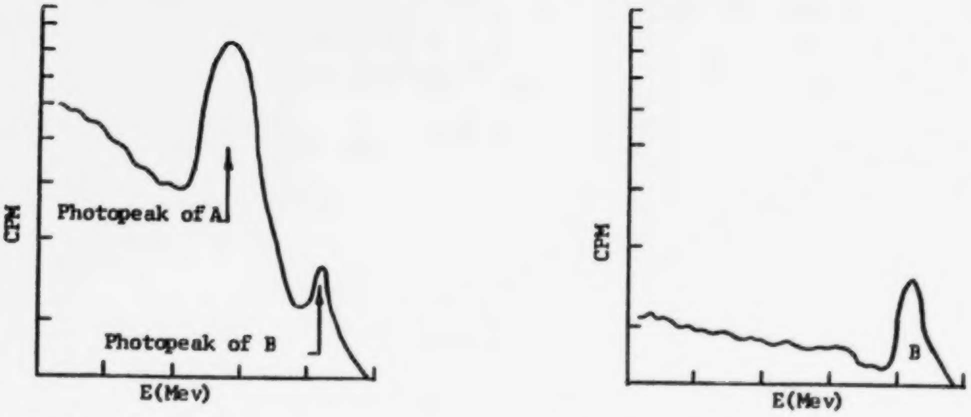
The repeatibility of the standardization (dilutions, source preparations, counting statistics, etc.) was such that the estimated activity had a STANDARD ERROR (σ) less than ± 0.5 %. The estimated MAXIMUM SYSTEMATIC ERROR (Sum of the estimated maximum errors due to dispensing, counting losses, counting corrections, etc.) was less than + three% (δ) and - three% (δ'). The TRUE ACTIVITY is therefore claimed to be within the limits of $+(3\sigma+\delta)$ and $-(3\sigma+\delta')$, that is + five% and - five% of the quoted value."

If a particular standard is found to be contaminated, reference to information such as that presented in Table 2 is helpful for identification purposes. Many of the possible contaminants that may interfere in a given calibration situation can be eliminated by energy, half-life and decay scheme considerations. Complete correction for impurities may not always be possible under present methods of production and separation. However, in cases where the impurity is identified or can be determined empirically, suitable corrections for the interference can be made by spectrum stripping methods. For example, an accurate efficiency value is desired for radionuclide A. A standard of A is obtained from a supplier but the accompanying certificate states that the standard contains a certain percent C of impurity B which interferes with the calibration.

Figure 1 presents one method by which an accurate efficiency value may be obtained for nuclide A when the standard is contaminated with nuclide B. In (a) the combined spectrum of standard A plus the impurity B is illustrated. The spectrum of B, at the same activity level as in (a), is presented in (b). The relative contributions of A and B to the total spectrum is shown in (c). By subtracting (b) from (a) the net spectrum (d) is obtained. Spectrum (d) is used to calculate an instrument efficiency for nuclide A.

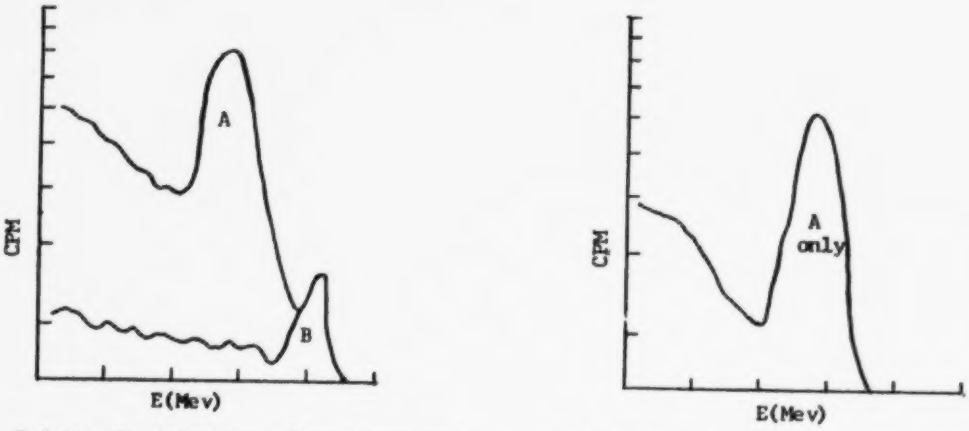
Table 2(1, 2): Activities associated with commonly used radionuclide standards

Isotope	Method of Production	Other Activities, Half-Life
Cs^{137}	U(n, f)____Cs^{137}	Cs^{134} (2.2 yrs)
I^{131}	$Te^{130}(n, \gamma)$ Te^{131}____I^{131}	Te^{121m} (154d), Te^{123m} (104d), Te^{125m} (58d), Te^{127m} (105d), Te^{129m} (41d), Te^{131m} (30 hr.), Xe^{131m} (12d)
I^{131}	U(n, f) Te^{131}____I^{131}.	negligible
Ba^{140}	U(n, f)____Ba^{140}	La^{140} (40.2 hr. daughter)
K^{40}	Natural occurring	negligible
Ce^{141}	$Ce^{140}(n, \gamma)$____Ce^{141}	Ce^{137} (35 hr.), La^{137} (6 X 10^4 yr.), Ce^{139} (140d), Ce^{143} (33.4 hr.), Pr^{143} (13.8d)
Ce^{141}	U(n, f)____Ce^{141}	Pr^{144} (17.3 min.), Ce^{144} (284d)
Ce^{144}	U(n, f)____Ce^{144}	Ce^{141} (32.5d), Pr^{144} (17.3 min.-daughter)
Zr^{95}	$Zr^{94}(n, \gamma)$____Zr^{95}	Zr^{97} (17 hr.), Nb^{97} (74 min.), Nb^{95} (35d) daughter Hf^{175} (70d), Hf^{181} (45.5d)
Zr^{95}	U(n, f)____Zr^{95}	Zr^{97} (17 hr.), Nb^{97} (24 min.), Nb^{95} (35d)
Ru^{103}	U(n, f)____Ru^{103}	Ru^{106} (40 days), Rh^{106} (30 sec.)
Ru^{106}	U(n, f)____Ru^{106}	Ru^{103} (40 d), Rh^{106} (30 sec.-daughter)
Co^{60}	$Co^{59}(n, \gamma)$____Co^{60}	Co^{60m} (10 min.)
Zn^{65}	$Zn^{64}(n, \gamma)$____Zn^{65}	Zn^{69m} (13.9 hr.), Zn^{69} (55 min.)
Cu^{64}	$Cu^{63}(n, \gamma)$____Cu^{64}	negligible



(a) Combined Spectra

(b) Spectrum of Impurity



(c) Relative Contributions of each Spectrum

(d) Net Spectrum

SUBTRACTION METHOD FOR OBTAINING ACCURATE EFFICIENCY VALUE

Figure 1

The subtraction method for removing impurities is accurate and quite useful for calibration purposes provided the contaminants and their quantities are known. Unfortunately many suppliers of standards do not furnish the information required for the treatment.

Because of the variation in the amount and type of information supplied in the certificate accompanying the standard, the Calibrations and Standards group of the Radiological Health Research Activities, Robert A. Taft Sanitary Engeering Center has proposed a model format for radionuclide standard certificates. The format along with an example of the proposed certificate for a radionuclide standard is reproduced below:

TABLE 3
INFORMATION REQUIREMENTS FOR MODEL CERTIFICATE(4)

1. Description
    - a. Principal radionuclide
    - b. Chemical form
    - c. Solvent
    - d. Carrier and content
    - e. Mass and specific gravity or volume
2. Standardization
    - a. Activity per mass or volume
    - b. Date and time
    - c. Activity of daughter
    - d. Method of standardization
3. Accuracy
    - a. Reproducibility error
    - b. Systematic error
    - c. Overall error
    - d. Confidence levels
4. Purity
    - a. Identification of impurities
    - b. Activity
    - c. Included or not included in principal activity
5. Assumptions
    - a. Decay scheme
    - b. Half-life
    - c. Equilibrium ratios
6. Production
    - a. Production method
    - b. Date of separation
7. Usable lifetime

TABLE 4

EXAMPLE OF PROPOSED CERTIFICATE FOR A RADIOACTIVITY STANDARD(4)

DESCRIPTION

Principal Activity - Cerium-141
Chemical Form - $Ce(NO_3)_3$
Solvent - 0.01 N HNO_3
Carrier content - 25 micrograms Ce/ml
Volume - 3.13 ml

STANDARDIZATION

The concentration of the principal radionuclide has been found to be 145 microcuries/milliliter as of 1200 hours EST December 15, 1962.

The concentration of the daughter nuclide ____none____ was found to be ______ microcuries/milliliter at the same time, which has/has not been included in the principal activity.

Method of Standardization: $4\pi\beta\lambda$ coincidence

ACCURACY

The reproducibility (σ) of the standardization (dilutions, counting statistics, preparation, etc.) is ± 1% at one standard deviation. The systematic error (SE) (dispensing losses, counting losses, and corrections, etc.) is less than +2% and -2%. The true activity ($\pm 3\sigma$ +SE) therefore lies between the limits of + 5% and - 5%.

PURITY

The principal radionuclide was accompanied by the following types and amounts of other activities:

1. Pa-233 less than 0.5% of the principal activity
2. Pr-143 less than 0.1% of the principal activity

The activity of these impurities has/has not been included in the principal activity stated above.

TABLE 4--Continued

ASSUMPTIONS

The standardization method was based on the following decay scheme assumptions:

100% β emission followed by coincident photons of 0.145 Mev representing 49% of the disintegrations. 30% β emission to ground state was assumed.

Estimated half-life of principal radionuclide: 32.5 days

PRODUCTION

Date of production method separation	Ce-140 (n, λ) Ce-141 December 2, 1962

NOTE: This preparation should not be used after 10 half-lives have expired.

It is believed that use of such a certificate would aid in (1) correcting the inconsistencies among suppliers in reporting sources of error in the activity statement, (2) require the suppliers to indicate significant radioactive impurities and (3) require the suppliers to furnish complete information on the standardization technique, assumptions concerning decay schemes and equilibrium conditions.

IV. PROPERTIES OF RADIONUCLIDE SOLUTIONS

A. Chemical and physical properties

1. STORAGE

Calibrated radionuclide standard solutions are prepared for storage and shipment by the supplier in flame sealed glass ampoules. Studies have shown(5) that several radionuclides adsorb on polyethylene containers and certain solvents, e.g., HCl, diffuse through the walls of the polyethylene containers.

Therefore, it is recommended that all dilutions and storage of radionuclides be performed in glass containers and that the use of polyethylene be avoided.

2. CHEMICAL PROPERTIES

Although it is true that the chemistry of a radionuclide is the same as its stable nuclide counterpart, it is necessary to observe special precautions when preparing radionuclides for storage. On a weight basis the amount of radionuclide present, even in micro- or millicurie amounts, is extremely small and precautions are necessary to prevent "plating out" or adsorption losses, even on glass. Other precautionary measures necessary in preparing or diluting radionuclide standard solutions are concerned with a knowledge of the chemistry of the radionuclide in question.(6)

Listed below are the compositions of several radionuclide solutions as used at the Oak Ridge National Laboratory's Isotopes Development Center.(6, 7) It is obvious from this list that the solvents, carriers, and/or complexing agents must be varied to fit the particular radionuclide in question.

3. PHYSICAL PROPERTIES

Brief mention should be made of the fact that physical properties, such as boiling point and melting point are applicable to both radionuclides and stable nuclides of an element.

TABLE 5(6)

COMPOSITION OF TYPICAL RADIONUCLIDE SOLUTIONS

RADIONUCLIDE	COMPOUND	SOLUTION
Cs^{137}	CsCl	1 $\underline{N}$ HCl ± 50%
I^{131}	NaI	Na_2SO_3 - NaOH
Ba^{140}-La^{140}	$BaCl_2$	1 $\underline{N}$ HCl ± 50%
Ce^{141}, Ce^{144}-Pr^{144}	$CeCl_3$	1 $\underline{N}$ HCl ± 50%
Zr^{95}-Nb^{95}	Oxalate complex	5% oxalic acid
Ru^{103}, Ru^{106}-Rh^{106}	$RuCl_3$	6 $\underline{N}$ HCl
Co^{60}	$CoCl_2$	1 $\underline{N}$ HCl ± 50%
Zn^{65}	$ZnCl_2$	1 $\underline{N}$ HCl ± 50%
Fe^{59}	$FeCl_3$	1 $\underline{N}$ HCl ± 50%

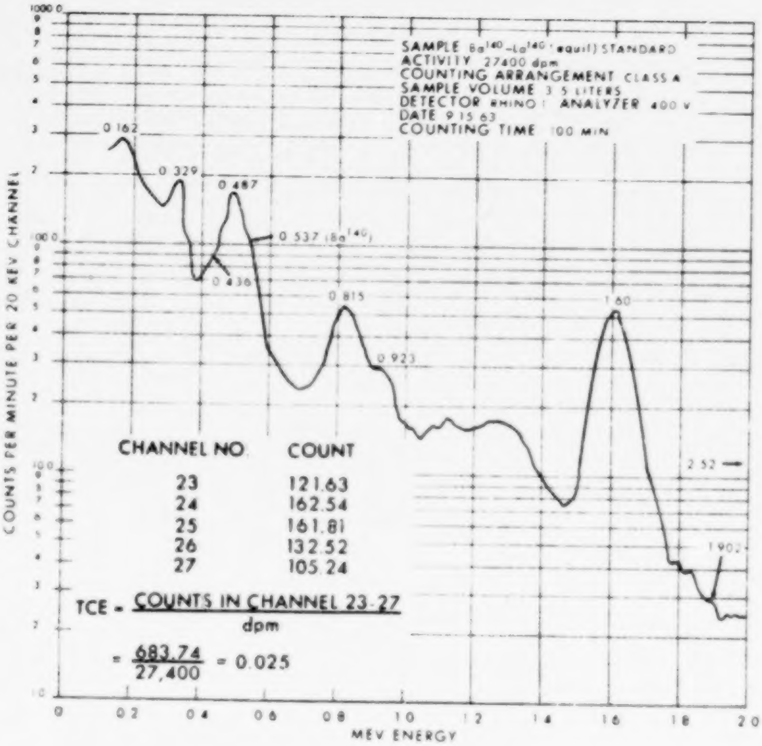
V. INSTRUMENT EFFICIENCY CALIBRATION

Instrument counting efficiency for a particular radionuclide should be strictly defined as to its application to counting data. Counting efficiencies are commonly described with reference to a particular radionuclide, spectrum area, decay scheme and counting arrangement. For most radionuclides of importance in environmental surveillance such a description is simple and straightforward. However, there are several calibration situations which require special consideration which reference to efficiency values.

A. Method for defining efficiency for special calibration situations

A standard spectrum of Ba^{140}-La^{140} at transient equilibrium is presented in Figure 2. Depending on how the Ba^{140}-La^{140} activity is defined, two values for the total counting efficiency may be obtained. The counting efficiency for the nuclide pair can be based on the total activity of the standard (i.e., Ba^{140} + La^{140} activity), or on just one member of the pair. The activity of either member of the decay chain is established by series decay relationships. Although both methods are equally correct, the total counting efficiency for series decay nuclides is often expressed in terms of both members. Thus the total photopeak counting efficiency (Figure 2) for Ba^{140}-La^{140} is 2.5 percent.

Another series decay pair of considerable interest is Zr^{95}-Nb^{95}. The total photopeak counting efficiency for this pair (Figure 3) is 2.2 percent. Of special interest in this case, and several others, is the fact that the



CHANNEL NO	COUNT
23	121.63
24	162.54
25	161.81
26	132.52
27	105.24

$$TCE = \frac{\text{COUNTS IN CHANNEL 23-27}}{\text{dpm}}$$

$$= \frac{683.74}{27,400} = 0.025$$

Figure 2

photopeak used for the efficiency calibration is composed of more than one distinct gamma energy. In cases where the gamma efficiency is desired for these radionuclides, the gamma abundance of each contributing photon must be considered in the calculation. The gamma efficiency for Zr^{95}-Nb^{95}, based on three gamma photons of different energies, would be calculated as shown in equation (1)

$$\text{eff} = \frac{\text{cpm}}{(\gamma_{ab_1} + \gamma_{ab_2} + \gamma_{ab_3})\,(\text{dpm})} \qquad (1)$$

where:

λ eff is one gamma efficiency and

λ_{ab} is the fractional gamma abundance

B. Estimating efficiency

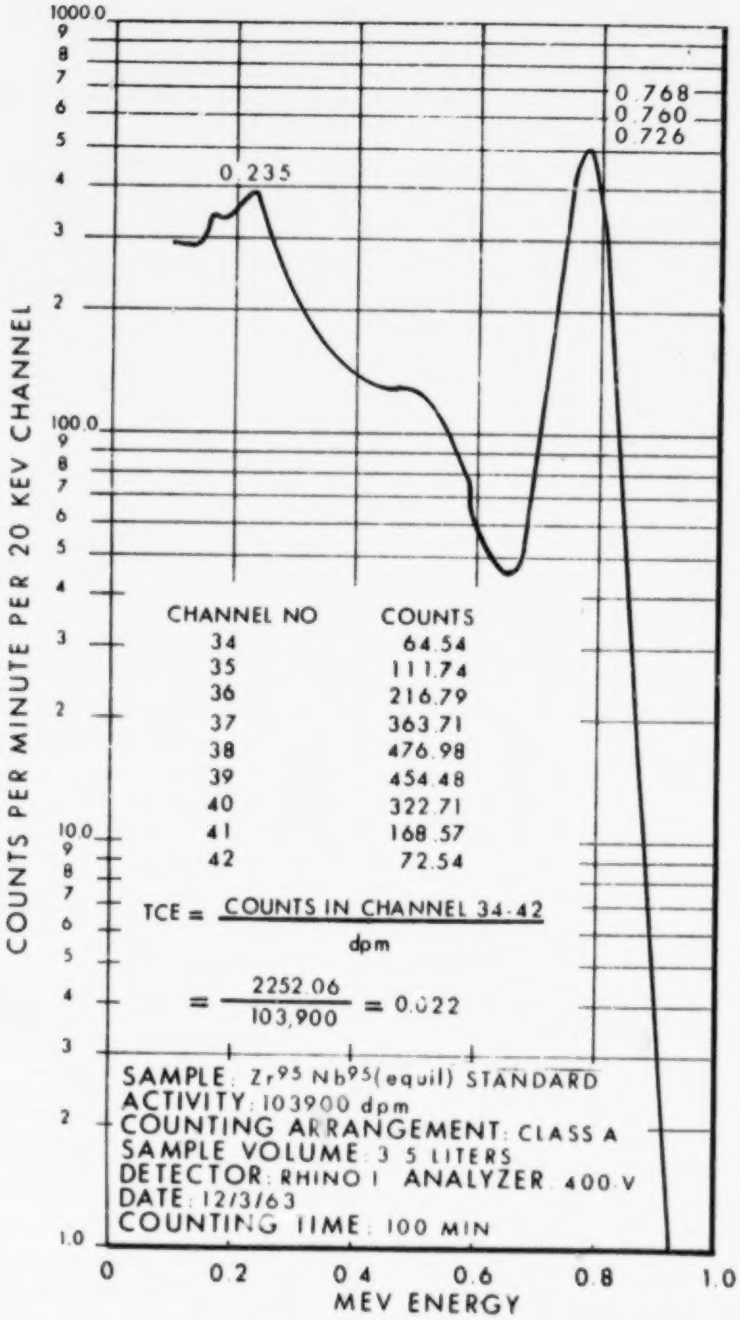
A useful tool for estimating gamma efficiencies is a plot on log-log paper of gamma efficiency versus energy obtained from the available radionuclide standards. Gamma efficiencies for other radionuclides may be obtained from this plot providing that a number of qualifications can be met.

1. The gamma efficiencies of standard radionuclides must be from single photopeaks, i.e., there must be no interference from higher energy photopeaks.

2. The gamma efficiency of standard radionuclides must be obtained from clearly defined photopeaks, i.e., not overlapping as in the case of Co^{60}.

3. The photopeak energy of radionuclides for which the efficiency is desired must also meet the above mentioned criteria therefore their decay schemes must be known.

In practice the gamma efficiency is usually converted to total counting efficiency and then used in subsequent calculations.



CHANNEL NO	COUNTS
34	64.54
35	111.74
36	216.79
37	363.71
38	476.98
39	454.48
40	322.71
41	168.57
42	72.54

$$\text{TCE} = \frac{\text{COUNTS IN CHANNEL 34-42}}{\text{dpm}}$$

$$= \frac{2252.06}{103{,}900} = 0.022$$

SAMPLE: Zr^{95} Nb^{95}(equil) STANDARD
ACTIVITY: 103900 dpm
COUNTING ARRANGEMENT: CLASS A
SAMPLE VOLUME: 3.5 LITERS
DETECTOR: RHINO I ANALYZER: 400 V
DATE: 12/3/63
COUNTING TIME: 100 MIN

Figure 3

VI. ACTIVITY LEVEL OF CALIBRATION

One of the first decisions to be made in the efficiency calibration is that of deciding the activity level to be used. Figure 4 shows the effect of activity level on instrument gain or channel shift for RHINO I, a detector system used by the Environmental Radiological Health Training Section of the Robert A. Taft Sanitary Engineering Center. The curve is representative of a very poor phototube. The curve of Figure 4 was obtained by counting Cs^{137} standards of increasing activity, under identical conditions of geometry, and observing the instrument channel shift. All the observations were normalized to a single base gain.

Channel shift at increasing activity levels is primarily related to a temporary increase in photomultiplier gain.(8) Since the phenomenon is a property of the instrument proper, the best method of evaluation is a plot similar to that of Figure 4. From such a plot and an evaluation of the expected range of activity levels to be encountered in the analysis, an activity level related to an acceptable channel shift can be determined. It should also be remembered that the amount of gain shift is dependent on the energy of the γ photon as well as the count rate.

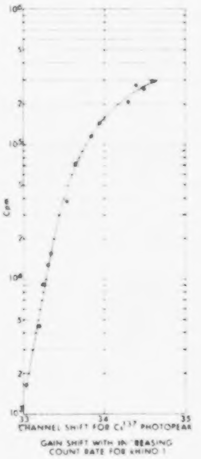


Figure 4

VII. SUMMARY

Care must be exercised in selecting and using radionuclide standards. A thorough knowledge of the chemical and physical properties of the radionuclide standard is necessary plus strict adherence to the information given on the radioactivity certificate, assuming the certificate gives sufficient information.

An accurate and well defined instrument efficiency is required for environmental surveillance work. Additional instrumental problems such as the activity level used in calibration procedures must be determined individually for each particular case.

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Preparation of Samples and Standards for Gamma Spectral Analysis

I. INTRODUCTION

One of the major considerations involved in choosing gamma spectroscopy over beta counting in the radionuclide analysis of environmental samples is the length of time necessary for preparing the samples for counting. A comparison invariably reveals a shorter length of time to prepare samples for gamma spectral analysis. However, the idea that no sample preparation is necessary for gamma spectral analysis is a misconception.

The preparation of samples for gamma spectral analysis is complicated by a number of factors. These factors include geometry, stability of sample, homogeneity of sample, complexity of the resulting gamma spectra, and calibration considerations involving all the foregoing factors.

Although it is customary to perform gross alpha and beta analyses on environmental samples such as air and water, it is not customary to perform gross gamma analyses. There is a wide variation in gamma photopeak counting efficiencies for various photon energies and also a wide variation in complexity of photon decay schemes. These variations make it extremely difficult to design a calibration standard for gross gamma analysis. Therefore, gross gamma analysis of environmental samples is not recommended, and this discussion will be confined to preparing samples for specific radionuclide analysis by gamma spectroscopy.

II. SAMPLE COUNTING ARRANGEMENT

The selection of the shape and size of a sample container will depend on the activity concentration and the physical size or volume of the sample.

A. Container Materials

The containers are usually made of glass, aluminum, stainless steel or plastic. When using glass, breakage and subsequent contamination is a problem. Also, impurities in the glass, such as potassium and radium, add to the undesirable background count. Glass has one desirable advantage in that it is relatively easy to clean.

Sample containers made of aluminum are frequently used. They are relatively easy to handle, inexpensive and are fairly free of radioactivity. However, aluminum is easily deformed and is attacked by strong acid solutions.

Stainless steel is another material widely used for sample containers. Although most steel made is contaminated with natural or artificial radioactivity, steel can be found that is relatively uncontaminated. Stainless steel is resistant to most reagents used in the laboratory but strong solutions of HC1 will react with it.

Plastic is another material that is becoming popular for use as sample containers. They are relatively inexpensive and easily decontaminated if they are molded or extruded in one piece. Some plastics which use metal powder as plasticizers or fillers may be contaminated with radium in the metal.

When considering the relative merits of different materials for use as sample containers, some thought should also be given to the feasibility of disposable containers. The expense of decontamination may exceed the cost of a container itself.

B. Container Configuration

The common types of sample containers are planchets, "cottage cheese type containers", and Marinelli beakers.

1. PLANCHETS

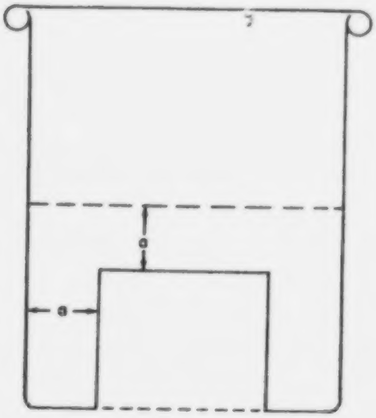
Planchets of the above materials may be obtained in flat or cupped shapes and are obtainable in various sizes. Planchets with concentric rings, raised ridges on the bottom surface, which assure better distribution of residue from evaporation, may also be obtained. Planchets are most advantageous for samples that may be evaporated. Petri dishes with tight fitting lids, such as those used with millipore filters, are useful for small volume liquid samples, i.e., less than 5 ml. The geometry of these is relatively high because of the closeness of the sample to the crystal.

2. COTTAGE CHEESE TYPE

Plastic cottage cheese containers, varying in volume from 50 ml to 1500 ml, are commercially available. They have the advantage of being water tight even when inverted. However, they are fairly brittle and are breakable. Caution should be exercised to reproduce their position when placing them on the crystal.

3. MARINELLI BEAKERS

In order to count bulky samples with better total efficiency than the cottage cheese type container, a Marinelli beaker is used. Marinelli beakers (see Figure 1) are special containers which have a cylindrical cavity in the center extending up into the beaker. The beaker is lowered over the crystal so the sample partially surrounds the crystal. This is analogous to the well type crystal, except now the sample to be counted is surrounding the crystal instead of the crystal surrounding the sample. The main advantage in using this arrangement is being able to count environmental samples without intermediate preparatory steps.



CROSS SECTION OF MARINELLI BEAKERS

Figure 1

4. FILTER PAPER

In some cases the final sample to be counted will be present on a filter paper. This occurs in air sampling where a large volume of air is passed through a filter paper or in chemical procedures when a residue is deposited on a filter paper.

Filter papers are then gamma scanned after folding the paper several times until a size corresponding approximately to the crystal face size is obtained. The paper can then be wrapped in tape and placed directly on the crystal or placed in some sort of plastic container for counting. Placing a weight on top of the filter paper tends to improve the reproducibility of geometry.

C. Sample Size

The selection of sample size involves two closely related properties, the activity concentration of the sample and the sample configuration which in turn will determine the counting efficiency. For high activity concentration samples, small volumes, 1 ml to 50 ml, can be used and still obtain the necessary counting statistics. It should be noted that relatively good counting efficiency will be obtained with these small volumes. For low activity concentration samples the problem becomes more complex. Sample volume must be enlarged to increase the number of disintegrations taking place within the sample. However, the counting efficiency will decrease as the average source to crystal distance increases. Fortunately, the net effect is an increase in the count rate. The actual selection of counting arrangement and sample volume will depend on the minimum sensitivity of the analysis required.

III. PREPARATION OF SAMPLES AND STANDARDS

After the samples are collected, it is necessary to prepare them for gamma spectral and/or radiochemical analysis. An attempt is made to count the sample either in its original form or in a form which alters the sample only to the point of producing a homogeneous sample. It is important that samples be thoroughly mixed since the total counting efficiency is directly dependent on the distance of the activity from the detector and, therefore, erroneous results can be obtained from non-uniform distribution of radioactivity in samples.

Calibration standards for determining counting efficiencies should be prepared in such a way that they are counted under conditions identical to the unknown samples. These conditions relate mainly to reproduction of geometry and density between standards and unknowns.

A. Liquid Samples

1. MILK

Homogeneity of milk samples is normally not a problem because formalin is routinely added as preservative to samples collected in the field and the samples remain stable during the period from collection to counting. (If the milk curdles or separates, there is an uncertainty as to whether the radionuclides concentrate in the separated solids or the liquid portion.) The usual procedure is to pour 3.5 liters of milk into a Marinelli beaker and gamma count the sample on a large sodium iodide crystal detector which is connected to a multi-channel analyzer.(2) The spectra obtained are complex, but since the principal gamma-emitting radionuclides that can be present in milk are I^{131}, Ba^{140}-La^{140}, Cs^{137}, and K^{40}, the concentration of each radionuclide can be calculated from a set of simultaneous equations.

Efficiencies for I^{131}, Cs^{137}, Ba^{140}-La^{140}, and K^{40} are determined by adding a known amount of each isotope to four different counting containers filled with distilled water to the required volume counting, and dividing the observed count rate by the disintegration rate of the activity added. Using these efficiencies, activity concentrations for these four radionuclides can be calculated from count rates determined by analysis of sample spectra.

2. WATER

Homogeneity and stability of water samples is normally not a problem except when suspended solid material is present. The solid material can be filtered out and treated as a separate sample.(2, 3) Samples such as tap, surface, well, and cistern water are normally counted in Marinelli beakers. Precipitation samples are usually counted in cottage cheese or smaller containers due to their higher specific activity or limited volume of available sample.

In the case of water samples, the calibration problem is not as well defined as for milk. The radionuclide compositions of tap, well, surface, saline, and rain waters are variable, and counting geometries also vary, depending on the quantity of available sample and the specific activity of the sample. If the radionuclide composition of the sample is known or can be determined, it is possible to set up and solve a series of simultaneous equations for these radionuclides.

B. Solid Samples

1. FOOD, VEGETATION, SOIL

Homogeneity of food, vegetation, and soil samples is a definite problem. It is necessary to homogenize food samples such as lettuce and potatoes, in a blender, garbage disposal unit or other type of grinder. After blending, the samples may be poured directly into a Marinelli beaker for counting, or the blended sample may be ashed and the resulting ash placed in a cottage cheese dish or other suitable container for counting.(4)

Vegetation samples are blended, if possible, and processed as food samples.

Soil samples are prepared for gamma counting by drying, and then grinding the dry material into a fine powder or by ashing as above. Many laboratories perform their efficiency and standard spectra calibrations using distilled water in plastic containers, and then apply these calibrations to ash samples. It should be pointed out that the density of inorganic ashes is lower than water, hence the counting efficiency will be different.

Where the volume of ash is small, the counting efficiencies between ash and water will not vary considerably, but for large volumes, e.g., 100 ml. or greater, the efficiencies will vary considerably.

If large volumes or weights of ash are to be gamma counted, it is suggested that standards be prepared which simulate the density of the ash. The Calibrations and Standards group of the Radiological Health Research Activities of the Robert A. Taft Sanitary Engineering Center has developed a method of calibration whereby sawdust is used to simulate the density of ash. Known amounts of standardized radionuclides are added to sawdust in a cottage cheese dish. Another empty cottage cheese dish is taped mouth to mouth to the dish containing the sawdust, and the sawdust is shaken back and forth between the dishes until the sawdust is thoroughly wetted with the radionuclide solution. The empty dish is then removed, the dish containing the sawdust is covered, and counted. Again, it is necessary to obtain standard spectra and counting efficiencies for every radionuclide for which activity concentration is to be determined.

2. AIR FILTERS

Homogeneity of air samples is very difficult to attain, due to the nature of the sampling process. The process involves filtering large volumes of air through a filter paper, usually a glass fiber paper.

In any case, calibration is very difficult to the geometry of the folded filter paper and the complexity of the spectra. Furthermore, since passing a standard radionuclide aerosol through a filter paper will not result in a calibration standard that is representative of environmental conditions, it is necessary to utilize other techniques.

One method of calibration used by Radiological Health Research Activities of Robert A. Taft Sanitary Engineering Center makes use of standard spectra obtained from the filters and comparison of these standard spectra to previously obtained efficiencies in another geometry, such as distilled water in a cottage cheese container. This is accomplished as follows:

(a) Obtain counting efficiences for the desired radionuclides in a cottage cheese container in a suitable volume of distilled water.

(b) Add an uncalibrated amount of a radionuclide, e.g., Cs^{137}, to a filter paper in such a volume that the sampling area of the filter is moistened.

(c) Dry the filter paper under a heat lamp.

(d) Fold the filter paper and count.

(e) Plot the standard spectra.

(f) Dissolve the filter paper in a minimum volume of HF, transfer to a cottage cheese dish and dilute to the volume as prescribed in (a).

(g) Count the dissolved paper and from the gamma counts observed, calculate the disintegration rate of Cs^{137} present in the sample, i.e., which was present on the filter paper.

(h) From the disintegration rate of Cs^{137} on the filter paper and the number of observed counts on the filter paper, calculate the counting efficiency.

(i) Repeat this procedure for every radionuclide of interest. It must be remembered, however, that this calibration procedure is dependent on the chemistry of the radionuclides involved. Not every radionuclide can be handled exactly like Cs^{137}. Some radionuclides, e.g., Ce^{141} and Ce^{144} are insoluble in HF and a different procedure must be employed for preparing and dissolving the standards.

An alternate method to calibrating for every radionuclide of interest is to calibrate for about three radionuclides of selected energies, and then construct a gamma photopeak efficiency versus energy curve. An approximate gamma photopeak efficiency for other radionuclides may then be obtained from this graph.

IV. SUMMARY

Sample preparation is an integral part of any gamma spectral analysis program. With information gained from experience and "trial" runs, the following factors need to be considered:

1. What will be the geometry of the sample?

2. Will the sample be homogeneous?

3. Will the sample be chemically and physically stable?

4. Will it be possible to calibrate for the expected geometric and spectral situation?

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GPO 869-164

Radionuclide Identification by Energy and Half-Life

I. INTRODUCTION

One of the basic uses of gamma ray spectroscopy is qualitative identification of gamma-emitting radionuclides in environmental samples. Fundamental to all schemes for this qualitative analysis is the initial attempt to determine the nuclides present by energy and half-life.

II. NUCLIDE IDENTIFICATION BY ENERGY

Realizing the possible complexities that may occur in a gamma spectrum, an organized procedure of analysis is required. A number of different techniques are used in practice and experienced personnel are able to apply many short cuts. The format given here, though only a suggested method, does include most of the essential elements.

A. Selection of Possible Nuclides on Basis of Photopeaks

The first step is usually to determine the energy of those peaks that appear in the spectrum and determine what nuclides may have contributed to them. The energy determination of the peaks is relatively simple if a good energy calibration has been made.

The selection of possible nuclides must take into consideration knowledge of the sample. The usual procedure is to assume that all peaks are photopeaks and then look up the nuclides with these gamma energies. The possible nuclides listed for a single peak should extend over an energy range that coincides with at least plus and minus one channel from the observed energy. This takes into consideration possible gain shifts in the instrumentation and also, to some extent, the possibility of more than one gamma transition contributing to the peak.

When listing the possibilities for each peak some other items of information about them should also be listed; i.e., all the gamma transitions of the nuclide, its half-life, and possibly its parent and daughter relationships (especially if they are also gamma emitters).

A number of tools have been found to be helpful in compiling such a list. On page 95 of the Radiological Health Handbook is a table of radionuclides arranged by gamma-ray energy which includes other useful information. The Chart of the Nuclides on page 90 and the Table of Isotopes starting on page 220 of the Radiological Health Handbook are also most helpful. A more comprehensive list of gamma ray information is given in the Table of Gamma-Rays Emitted by Radioactive Nuclei, Arranged in Order of Increasing Energy, compiled by E. der Mateosian and M. McKeown of Brookhaven National Laboratory and published as report number B NL-605 (T-177). This report is published by the Office of Technical Services, Department of Commerce, Washington, D.C.

B. Analysis of Peaks Other Than Photopeaks

The second step might be an analysis of peaks that are suspected not to be photopeaks. For example, some of the lower energy peaks may well be backscatter peaks or escape peaks due to higher energy gammas. These may be checked to see if they could have resulted from any of the higher energy photopeaks that are present. Peaks at 0.51 and 1.02 Mev may indicate positron emitters and these may need to be listed as possibilities. Characteristic x-rays may show up as peaks at lower energies and these should be checked. The Table of Electron Binding Energies which starts on page 212 of the Radiological Health Handbook can be used in analyzing for the possibility of characteristic x-rays. The most usual characteristic x-ray of an element is found at an energy equal to the binding energy difference between K and L shells.

C. Comparison of Unknown Spectrum with Standard Spectra of Possibilities

A third step is to compare, for points of similarity, the spectrum of the unidentified nuclides and the standard spectra of the possibilities. Several publications have catalogues of standard spectra, among these are the listed references 2, 3, and 4. A catalogue of standard spectra obtained on the same spectrometer as the sample spectrum is most useful for its comparison. The relative peak heights and the peak energies of the Standard Spectra should be similar to the unknown if due to the same single nuclide. Care must be exercised, however, as more than one nuclide may be contributing to a peak, thus causing the relative peak heights to be greatly different.

D. Compilation of Final List of Possibilities

After making a complete analysis, as mentioned above, a number of the original possibilities can probably be eliminated. This elimination may be on the basis of peaks not showing up for other gamma transitions, absences of daughter components that should appear, or other circumstances. The remaining possibilities are the best estimate of the gamma-emitting nuclide composition that can be made up to this point.

III. RADIONUCLIDE IDENTIFICATION BY HALF-LIFE

The next step in analysis is to make subsequent counts at later time periods. The resulting spectra will lend themselves to further analysis. After a number of spectra at later time periods have been determined, it may be obvious that certain peaks are showing up that were not seen before. This is due to the decay of shorter half-life materials whose spectra were hiding these peaks.

If such is the case, an energy analysis of these peaks should also be conducted.

One of the greatest benefits that can be obtained from spectra made at different times is a determination of the half-lives of the nuclides that are contributing to the photopeaks. The radioactive decay law may be expressed by the simple equation:

where:

$$A_2 = A_1 e^{-\lambda \Delta t} \qquad (3)$$

A_1 = the original activity at time t_1

A_2 = the activity at a later time t_2

e = the base of the natural logarithm

λ = the decay constant = $0.693/T_{\frac{1}{2}}$

where $T_{\frac{1}{2}}$ is the half-life

$\Delta t = t_2 - t_1$

By having quantities which are proportional to A_1 and A_2, and knowing the appropriate times, t_1 and t_2, we may evaluate the half-life ($T_{\frac{1}{2}}$).

A. Determination of a Quantity Proportional to Activity

If only one single photopeak existed in the spectrum, with no higher energy gammas contributing any counts to this photopeak area, the total height of the photopeak could be used as a quantity proportional to the activity. However, this is very seldom the case, and an allowance must be made for the fact that other gammas, through Compton interaction and other means, are contributing to the photopeak that is being analyzed. In order to make an approximate subtraction of the Compton Continuum, a line is drawn along

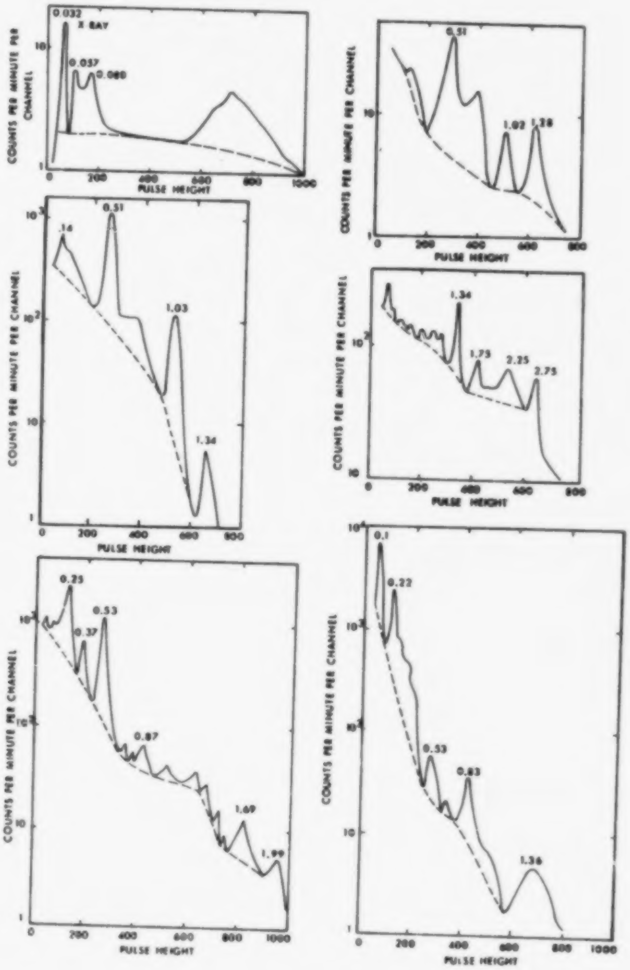


Figure 1

Example of Compton Continuum line

the base of the photopeaks. There is no precise "right" way to draw this Compton Continuum line. It is largely a matter of practice and judgment. Figure 1 shows by dashed lines several examples of Compton Continuum lines.

The relative activity is then determined for each counting time by subtracting the count rate value of the Compton Continuum line at the same energy as the photopeak from the count rate value of the photopeak. This procedure is illustrated in Example 1.

Example 1:

For the spectrum in Figure 2 determine the relative activity for the 1.54 Mev peak at each of the counting times.

Solution:

8 days after sample collection

0 days after 1st counting time

1.54 Mev Peak height	140
Compton Continuum line height	-65
Relative activity	75

13 days after sample collection

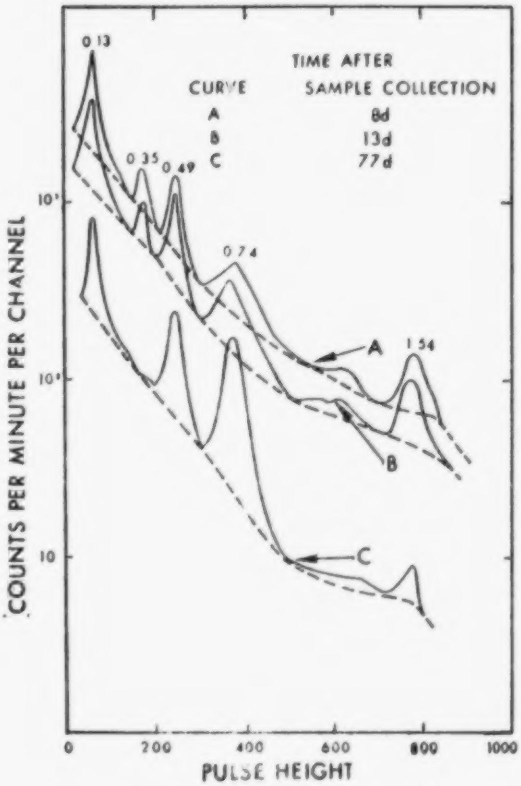
5 days after 1st counting time

1.54 Mev Peak height	100
Compton Continuum line height	-42
Relative activity	58

77 days after sample collection

69 days after 1st counting time

1.54 Mev Peak height	9.0
Compton Continuum line height	-5.5
Relative activity	3.5



GAMMA SPECTRUM OF AIR SAMPLE
COUNTED AT THREE DIFFERENT TIMES

Figure 2

B. Determination of Half-Life

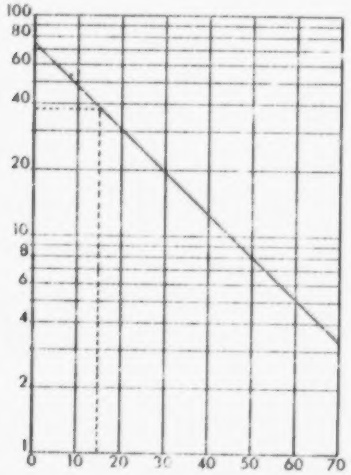
By having a relative activity term for a peak, for each counting period, these values could be substituted for activities in the basic decay equation (3) with the appropriate t values, and $T_{\frac{1}{2}}$ determined. It is often simpler, however, to plot the log of the relative activity vs. time and pick off the time required for the activity to decrease by one-half. This time is then the half-life of the peak analyzed. This latter technique is shown in Example 2.

Example 2

Determine the half-life for the 1.54 Mev peak of Figure 2.

Figure 3 is a semi-log plot of relative activity vs. time after the first counting time.

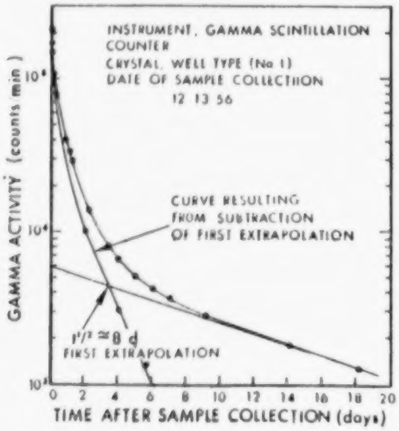
The original relative activity was 75. The point where half the activity has gone (i.e., $\frac{75}{2}$ 37.5 relative activity) is at [illegible] 15 days. This is the best estimate of the 1.54 Mev peak's half-life.



HALF-LIFE DETERMINATION FOR EXAMPLE 1

Figure 3

Having a value of the half-life for a peak, even though approximate, allows one to rule out many of the remaining possibilities from the energy analysis. Often it may be possible, at this point, to conclusively identify some of the nuclides in the spectrum of unidentified nuclides.



RESULT OF HALF-LIFE DETERMINATION FOR PEAK WITH MORE THAN ONE CONTRIBUTING NUCLIDE

Figure 4

The determination of the half-life of a peak does not always work out as easily as shown in Example 2. It may be that more than one nuclide is contributing to the peak. If the half-lives of the nuclides are close together, it may be impossible to tell that there is more than one by this technique. If the nuclides are quite different in half-lives, the resulting semi-log plot may be a curved line such as shown in Figure 4. In this case, the lower part of the curve may be extrapolated back, as shown in Figure 4, to obtain a line representing the plot for the longer-lived nuclide. The values of this extrapolated line can then be subtracted from the curve, giving the plot for the shorter half-life materials, as shown in Figure 4. There may be a 3rd nuclide with a still shorter half-life, as appears to be the case with Figure 4, for a curved line is still observed. To resolve the situation further, the lower end of this curve would be extrapolated and a second subtraction made. The accuracy of this technique is somewhat in question after the third extrapolation, especially if the half-lives are fairly close to each other.

Another complication arises when ingrowth of a gamma emitting daughter is occurring in the sample. In this case, the relative activities at subsequent counting times may appear to increase, to stay about the same, or to decrease, depending on the type of series decay and the degree of equilibrium attained.

IV. DIFFERENCE SPECTRA

Another technique that is useful in qualitative spectrum analysis, when working with spectra made at subsequent time intervals, is that of analysis of difference spectra. Difference spectra are obtained by the subtraction of one spectrum from the spectrum at some previous counting time. The result is a spectrum of the activity that has decayed in the interim time. Quite often, peaks show up in this "difference spectrum" that were not seen before. This technique becomes quite useful when some nuclides with short half-lives are present, as they often show up quite well in the difference spectrum.

V. SUMMARY

The analysis of a gamma-ray spectrum for qualitative identification of the gamma emitting components usually include an analysis of the energies of the peaks and the half-lives of the peaks. Both techniques are very useful and indeed, in some cases, give conclusive evidence as to the nuclides contributing to the spectrum.

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Radionuclide Composition of Environmental Samples

I. INTRODUCTION

Efficient qualitative and quantitative analysis by gamma spectroscopy necessitates a knowledge of what radionuclides should be expected for given types of environmental samples. In order to evaluate the more biologically significant radionuclides, the total radionuclide composition of the sample must be known.

This outline will discuss the mechanisms of production of radionuclides and the expected radionuclide composition of environmental samples when contaminated by various sources.

II. MECHANISMS OF PRODUCTION

The significant radionuclides in the environment are neutron induced fission products, neutron activation products, and naturally radioactive nuclides. Although some radionuclides are produced by such processes as proton activation, they are not a general environmental problem and so will not be considered here.

A. Neutron Induced Fission Products

Fissioning is a process in which a heavy nucleus breaks up into two roughly equal weight nuclei. The majority of the newly formed nuclides, termed primary fission products, are radioactive and most of these undergo a series of decays giving rise to a large number of secondary fission products. There are over thirty modes in which the uranium-235 nucleus can undergo neutron induced fission. There are over sixty primary fission products and the decay chain products total about two hundred nuclides. As an example, if one atom of uranium-235 undergoes neutron induced fission, it can yield yttrium-95, iodine-138, two neutrons, and fission energy. Typical fission yield curves for the neutron induced fissioning of uranium-235, uranium-238, and plutonium-239 are shown in Figure 1.

Not all of the fission products can be analyzed by gamma spectroscopy. Strontium-90, the most hazardous fission product, is a pure beta emitter which decays to yttrium-90 and then to stable zirconium-90. While yttrium-90 does emit a gamma photon,

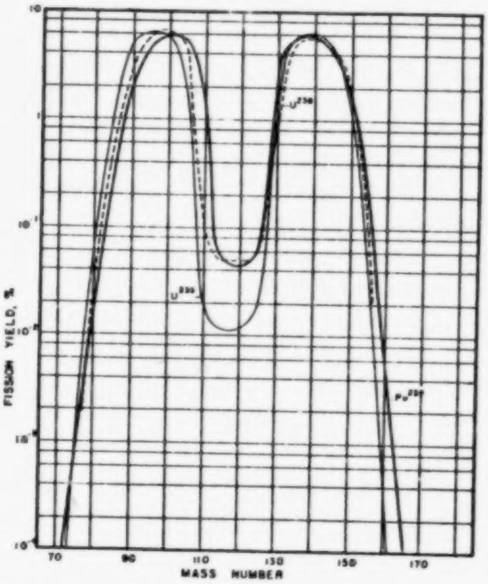


Figure 1. Fission yields for slow-neutron fission of U^{235}, U^{238}, and Pu^{239} versus mass number(1)

its low gamma abundance makes its use unfeasible for gamma spectral analysis of environmental samples. Other fission products also may be pure beta emitters or have gamma abundances too low for gamma spectral analysis.

B. Neutron activation products

Atoms of most elements, when exposed to slow neutrons, capture them to form a radioactive species of the same element. The probability of neutron activation or cross-section (susceptibility) differs for the various stable nuclides. The neutron activation of construction materials of the reactor or weapon, coolant water or air, water or air impurities, and surroundings such as the earth in surface weapons tests can produce considerable quantities of radionuclides. These radionuclides may then contaminate the environment.

C. Natural radioactivity

Some naturally radioactive materials were produced during the formation of the earth. Those with sufficiently long half-lives exist today. Other naturally radioactive materials continue to be produced by cosmic ray interactions. Members of the heavy element decay series and potassium-40 are examples of the first class whereas carbon-14 and tritium are examples of the second class. The identification and quantification of natural radionuclides is generally difficult due to their low levels. An additional problem is their interference in the determination of man-made radionuclides.

III. SOURCES OF CONTAMINATION

A number of sources of environmental contamination will be surveyed to determine what radionuclides one would expect to find in appropriate environmental samples.

A. Normal reactor operations

A reactor is an apparatus in which nuclear fission is sustained in a self-supporting chain reaction. It includes fissionable material (fuel) such as uranium, moderating material to thermalize neutrons (except for a fast reactor), and a reflector to decrease the number of neutrons escaping from the fuel. Provision is made for heat removal, control elements, and monitoring instrumentation.

1. SINGLE PASS AIR COOLED REACTOR

During normal operation, single pass air-cooled reactors put into the atmosphere significant quantities of argon-41 (activation of argon-40) and iodine-131 fission products. Particulate matter may undergo activation also. As an example, the Brookhaven Graphite Research Reactor (BGRR) puts out about 16,000 curies per day of argon-41.(2) This activity is introduced into the atmosphere by a 400 foot high stack and is diluted with ambient air. About 10 millicuries per day of iodine-131 is also released to the atmosphere.(3) Liquid wastes are representative of other installations and will be discussed later.

2. SINGLE PASS WATER COOLED REACTOR

The single pass water-cooled reactors in this country are the plutonium production reactors at Hanford, Washington. Cooling water for the reactors is taken from the Columbia River, subjected to water treatment processes to remove suspended impurities, passed through the reactors, held for several hours for decay of short-lived radionuclides, and returned to the Columbia River. The activity in the effluent is produced mainly by neutron activation, but also by fissioning of the uranium in the coolant waters and by leakage of fission products from failing fuel element jackets.

The radionuclides which have been identified are given in Table 1.(4)

Table 1. Relative abundance of reactor effluent radionuclides*

90% of total activity	8% of total activity	2% of total activity		
Na^{24}	P^{32} +	C^{14}	Ca^{45}	Sc^{46}
Si^{31}	Zn^{65}	Sc^{47}	Mn^{54}	Fe^{59}
Cr^{51}	Zn^{69}	Co^{60}	Sr^{85}	Sr^{89}
Mn^{56}	Ga^{72}	Sr^{90}	Y^{91}	Mo^{99}
Cu^{64}	Y^{90}	Ru^{103}	Zr^{95}	Cd^{115}
As^{76}	Sr^{91}	I^{131}	Ag^{111}	Cs^{137}
Np^{239}	Sr^{92}	Ba^{140}	I^{132}	La^{140}
	Y^{92}	Pr^{142}	La^{141}	Ce^{141}
	Y^{93}	$Ce\text{-}Pr^{144}$	Ce^{143}	Pr^{143}
	Nb^{97}	Nd^{147}	Pr^{145}	Pm^{147}
	I^{133}	Pm^{151}	Nd^{149}	Pm^{149}
	I^{135}	Sm^{156}	Eu^{152}	Sm^{153}
	U^{239}	Tb^{160}	Eu^{156}	Eu^{157}
		Ac^{227}	W^{187}	Po^{210}
			U^{238}	Pu^{239}

* Reference Time - 4 hours after irradiation

+ Radionuclides are underlined when their gamma energy is less than 0.1 Mev and/or less than a 5% gamma abundance.

As would be expected, most of the radionuclides have relatively short half-lives. These radionuclides would not necessarily be representative of a closed loop reactor since the longer half-life radionuclides predominate as time increases after irradiation. In addition to decay, some radionuclides will be preferentially deposited and therefore not carried downstream. Table 2 shows the relative abundances of various activities in the Hanford cooling water at several distances downstream.(5)

3. RECIRCULATING WATER COOLED

The radioactivity of concern in a recirculating or closed loop water reactor comes from activation of corrosion products, fission products leaking from defective fuel elements, or fission products resulting from fissioning of uranium contaminating the outside surfaces of the fuel elements. Under normal conditions the activity will be released from the demineralizer's ion-exchange resins, from primary coolant leakage, and from maintenance and decontamination operations.

Table 2. Annual average concentration of several radionuclides in Columbia River water - 1961

Units of pc/l

Radionuclide	7 Miles	40 Miles	260 Miles
Total Beta	56,000	11,000	520
Cu^{64}	15,000	2,200	ND
Mn^{56}	13,000	ND	ND
Cr^{51}	9,800	5,700	2,100
Na^{24}	9,500	1,800	ND*
Np^{239}	5,000	2,400	110
As^{76}	3,700	1,200	ND
RE + Y	2,000	390	24
Si^{31}	1,000	ND	ND
Ga^{72}	840	150	ND
Zn^{69m}	790	130	ND
Zn^{65}	600	340	90
P^{32}	410	260	68
Sc^{46}	57	33	24
I^{131}	18	10	ND
Sr^{89-90}	7.0	5.0	2.7
Sr^{90}	0.5	0.4	0.4

*Not Detected

Tritium may also be produced in large quantities as at Shippingport(6) where LiOH is added to the coolant water to raise the pH to 0.5 - 10.5. The higher pH reduces corrosion of the structurals and therefore reduces the amount of activation product formed, excepting tritium. About 50 curies of tritium were discharged to the river during the first year of operation.(7)

Actual operational experience with the nuclear powered naval ships and estimates for the N.S. Savannah indicate the following radionuclides may be present in appreciable quantities during normal operations.(8) The reactor coolant water will contain cobalt-60, iron-55, iron-59, tantalum-182, and chromium-51 as the important corrosion products. The coolant water will contain strontium-90, zirconium-95, ruthenium-106, cesium-137, niobium-95, and cerium-144 as important fission products.

The reactor coolant water of the U.S.S. Nautilus contains the following radionuclides in quantity;[9] cobalt-60, iron-59, chromium-51, tantalum-182, copper-64, iodine-131, strontium-90, cerium-144, cesium-137.

The spent demineralizer from the Nautilus contains:

Cobalt-60	10 curies
Cobalt-58	0.5 curies
Iron-59	0.5 curies
Chromium-51	0.3 curies
Manganese-54	0.2 curies
Hafnium-175	1.0 curies

The radionuclide differences are due to differences in materials of construction and fuel cladding.

The waste from a particular reactor may differ from that indicated in the examples to some extent. Since the materials suitable for construction of reactors are restricted, the examples should provide a good guide.

The operation of a water reactor will also produce amounts of volatile radionuclides. If these gases are collected and stored for a period of about 2 months, the 10 yr. krypton-85 nuclide will account for most of the activity remaining. It is then released to the atmosphere by stack discharge and diluted.

B. Reactor Accidents

The makeup of a radionuclide release in an accident depends upon the type of accident and the fission product inventory of the reactor at the time of the accident. Figures 2 and 2a[10] give the relative activities of a number of fission products with time. These are calculated and would be applicable to initial reactor startup, a criticality incident, or a nuclear detonation.

An operating reactor however, will have reached an equilibrium with the short half-life nuclides and will have a larger portion of the activity as longer half-life products. The fission product inventory in the core of a 500 megawatt reactor after 180 days of operation has been calculated[11] for 24 hours after shutdown and is summarized in Table 3.

There are great differences in the diffusion rates of the radionuclides into the water from the fuel elements. The halogens (bromine and iodine), the inert gases (xenon and krypton), and the alkali metals (cesium and rubidium) have escape rate coefficients about thirteen hundred times as great as the alkaline earths (strontium and barium).[12, 13] One would expect to find these elements escaping into the environment preferentially in case of an accident.

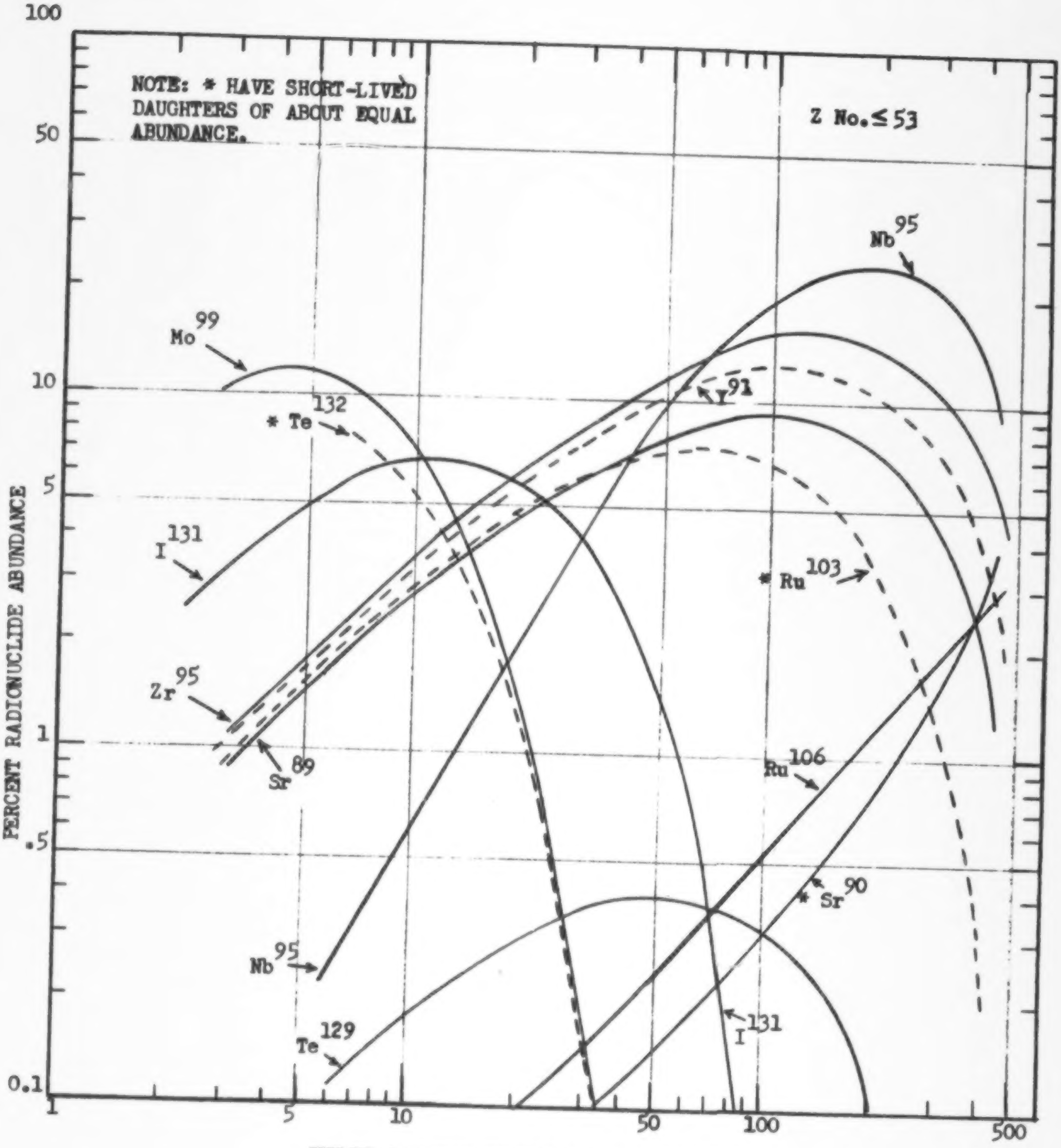
C. Fuel reprocessing

The nature of the radionuclides in fuel to be reprocessed can be approximated by the core inventory after a period of operation as in Table 3. Most of the wastes are contained and do not reach the environment. However, some do, particularly the gaseous and volatile radionuclides. Table 4[14] gives the average emission rates of several radionuclides from the separations plant stacks at Hanford during 1961.

Earlier measurements at Hanford have indicated that the rare earth and yttrium activity released is about 0.02 curies per day, strontium-89 activity released is about 0.004 curies per day and strontium-90 activity is about 0.0005 curies per day.

Most of the activity released to the water at Hanford comes from the production reactors discussed earlier. Oak Ridge also does separations work and the measurable amounts of activity discharged to the Clinch River for the year 1961 is given in Table 5.[15]

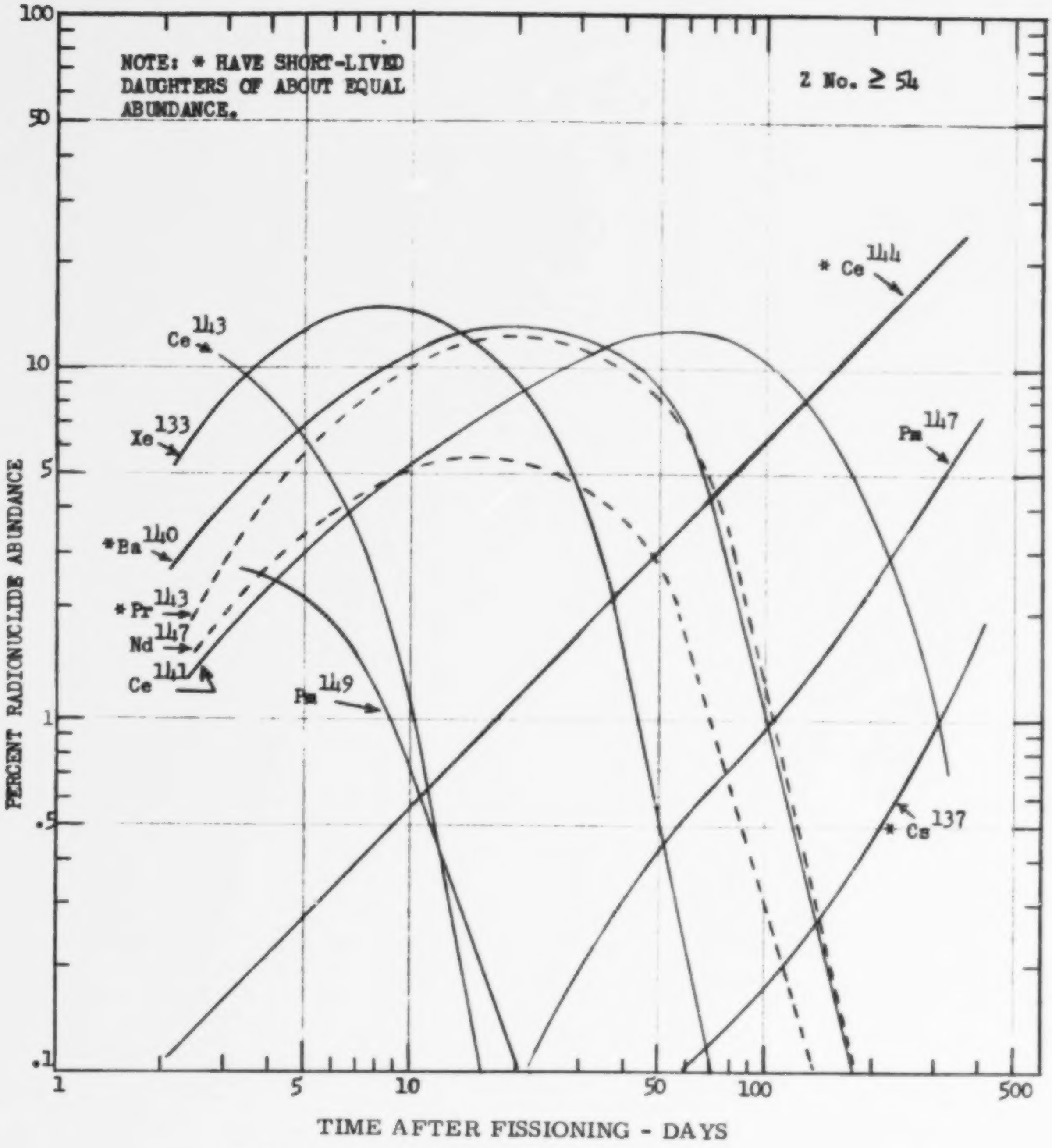
Other radionuclides which have been identified in the Clinch River and/or below the Savannah River plant by the Cooperative Studies Group, Radiological Health Research Activities[16] at the Robert A. Taft Sanitary Engineering Center include cesium-134, chromium-51, iron-59, and natural potassium-40.



TIME AFTER FISSIONING - DAYS

Relative Abundances of Fission Product Nuclides
Up to 500 Days After Fissioning

Figure 2



TIME AFTER FISSIONING - DAYS

Relative Abundances of Fission Product Nuclides
Up to 500 Days After Fissioning

Figure 2a

Table 3. 500 Mw reactor inventory - 180 day operation

Activity	Curies
Total	4.1×10^8
Volatile	8.4×10^7
Iodine	5×10^7
Noble Gases	3.4×10^7
Sr^{89}	1.7×10^7
Sr^{90}	3×10^5
Ce^{144}	8×10^6

Table 4. Average Hanford stack emissions - 1961

Radionuclide	Curies/day
I^{131}	0.7
$Zr\text{-}Nb^{95}$	0.005
Ru^{103}	0.003
Ru^{106}	0.005
Ce^{141}	0.006
Ce^{144}	0.01

D. Ore operations

The radionuclides from ore mining, milling, and processing operations are members of the uranium-238 and uranium-235 decay series. The most hazardous nuclide is radium-226.

Table 5. Activity discharged to Clinch River - 1961

Radionuclide	Curies/1961
Gross Beta	2230
Ru^{106}	2000
Cs^{137}	18
Sr^{90}	22
Co^{60}	31
I^{131}	3.7
Zr^{95}	20
Nb^{95}	70
Ce^{144}	4.2
Total RE Less Ce	24

E. Fallout

The nature of the radionuclides associated with fallout can be deduced from earlier discussions. The figures showing theoretical production (Figures 2 and 2a) apply to fallout for the fission products formed. The nature of the induced activities in fallout will depend mainly on the site of detonation. An atmospheric burst produces quantities of carbon-14 and tritium. Bomb structurals and the type of terrain exposed in a surface burst will determine the nature of the other induced activities. Typical radionuclides formed include sodium-24, silicon-31, aluminum-28, chlorine-38, cobalt-60, zinc-65, manganese-54, and scandium-46. Special activities such as tungsten-185 may be introduced as tracers. The High Altitude Sampling Program(16, 17) has resulted in identification of a large number of radionuclides in the upper atmosphere. Klement(18, 19) and others have reviewed the potential radionuclides that may be produced in weapons detonations.

F. Medical uses

Routine diagnostic and therapeutic applications of radionuclides are increasing in hospitals and medical research centers. Some radionuclides currently used internally(21, 22, 23) are listed in Table 6.

Table 6. Common medically used radionuclides

$\underline{H^{3*}}$	$\underline{C^{14}}$	Na^{22}	Na^{24}
$\underline{P^{32}}$	$\underline{S^{35}}$	$\underline{K^{42}}$	$\underline{Ca^{45}}$
Cr^{51}	Mn^{54}	Fe^{59}	Co^{60}
Cu^{64}	Zn^{65}	As^{74}	Br^{82}
$\underline{Y^{90}}$	Sb^{124}	I^{131}	Au^{198}

* Radionuclides are underlined which are not gamma emitters.

A number of radionuclides are used in sealed source form. Some common gamma emitters are radium-226, radon-222, cobalt-60, cesium-137, iridium-192, europium-152-154, ruthenium-106, and cerium-144. Some non-gamma emitters include strontium-90, yttrium-90, and phosphorus-32.

The common method of disposal of low-levels of these nuclides is by discharge to the sewerage system.

G. Natural

A great number of naturally occurring radionuclides may be present. The heavy element radioactive families(24) decaying from uranium-238, uranium-235, and thorium-232 provide much of the natural radioactivity. Other common naturally occurring radionuclides include tritium, carbon-14, and potassium-40.

IV. VECTOR CONTAMINATION

A. Air

The naturally occurring radionuclides in air generally include radon and thoron and their daughters.

Induced activity in air is contributed mainly by neutron activation to form carbon-14, tritium, and argon-41.

The fission products that can occur in air are limited only by the source of contamination, planned or accidental release into the air, and age after injection. Identification and evaluation efforts can be aided by use of the above information in a process of elimination.

Radionuclides analyzed for by surveillance networks include strontium-89, strontium-90, zirconium-95, cesium-137, barium-140, yttrium-91, cerium-141, cerium-144, promethium-147, and lead-210. Krypton-85 and iodine-131 are routinely released by operating reactors.

B. Fresh water

The predominant naturally occurring radionuclides in water include uranium-238, radium-226, thorium-232, potassium-40, carbon-14, and tritium.

Neutron activation products and fission products from reactor and fuel reprocessing operations have been discussed.

C. Sea water

The naturally occurring radionuclides in sea water should approximate those in fresh water. The use of nuclear powered ships and the use of the ocean bottom as a low-level disposal area will contribute neutron activation and fission products to the sea in addition to those of fallout from weapons testing.

The physical state of fission product elements after vaporization and injection into sea water has been studied by Greendale and Ballou.(25) Their data is given in Table 7.

D. Stream and marine biota

Radionuclide uptake by fish in the Columbia River has been studied extensively at Hanford. The surveillance program routinely analyzes fish for total beta, phosphorus-32, zinc-65, scandium-46, arsenic-76, chromium-51, and neptunium-239.

Table 7. Probable physical states of elements in sea water

Element	Ionic	Colloidal	Particulate
Strontium	87	3	10
Yttrium	0	4	96
Zirconium	1	3	96
Niobium	0	0	100
Molybdenum	30	10	60
Ruthenium	0	5	95
Antimony	73	15	12
Tellurium	45	43	12
Iodine	90	8	2
Cerium	2	4	94

Phosphorus-32, chromium-51, zinc-65, and neptunium-239 reached the Pacific Ocean in quantity. Oysters are routinely analyzed for scandium-46, potassium-40, zinc-65, zirconium-95, niobium-95, cesium-137, ruthenium-103, ruthenium-106, iodine-131, chromium-51, cerium-141, cerium-144, and phosphorus-32. Concentrations of zinc-65(26) were found which are in agreement with other studies.(27, 28) Other radionuclides which can concentrate in marine biota include cobalt-60 and iron-59 in body tissue, and cesium-137 in muscle. The experience in the Marshall Islands has provided considerable practical uptake data.(29) A number of radionuclides have been identified in fish as a result of nuclear testing.(30) These include manganese-54, iron-55, iron-59, cobalt-57, cobalt-58, cobalt-60, zinc-65, strontium-90, yttrium-90, cadmium-113m, cadmium-115m, cesium-137, barium-140, lanthanum-140, cerium-144, and promethium-144.

E. Milk

The predominant radionuclides(31) biologically and quantitatively in milk are potassium-40, iodine-131, barium-140, lanthanum-140, cesium-137, and strontium-90. Other radionuclides which may be present include radium-226, zinc-65, phosphorus-32, strontium-89, cobalt-60, iron-55, iron-59, tellurium-132, and carbon-14.

F. Meat

For radionuclides to appear in meat, they must be absorbed from the gastro intestinal tract of the animal. Radionuclides which are not absorbed include zirconium-95, niobium-95, cerium-141, promethium-147, other rare earths, and plutonium.(32) Iodine is taken up by the thyroid. It has been identified in ground beef.

Some radionuclides which are partly absorbed include iron-55, iron-59, cobalt-60, zinc-65, tellurium-132, and barium-140. Cesium is efficiently absorbed and would be present in the meat. Strontium is absorbed also but is a bone-seeker. Phosphorus-32 has been detected in ducks around Hanford.(33)

G. Vegetation

Although foliar deposition of any radionuclide could occur, the Hanford surveillance program routinely analyzes native grasses for barium-140, lanthanum-140, zirconium-95, niobium-95, ruthenium-103, ruthenium-106, iodine-131, cerium-141, and cerium-144. Pine trees around the waste pits at Oak Ridge have been sampled.(34) The identified radionuclides come from fallout, blowoff from the waste pits or seepage from the pits. They are ruthenium-106, zirconium-95, niobium-95, cesium-137, and cerium-144. Irrigated alfalfa around Hanford has been analyzed(35) and scandium-46, potassium-40, zinc-65, zirconium-95, niobium-95, cesium-137, ruthenium-103, ruthenium-106, iodine-131, chromium-51, phosphorus-32, strontium-89, and strontium-90 have been identified.

In analyzing for various types of vegetables around Hanford, only potassium-40, and strontium-90 appeared regularly. Zinc-65, and phosphorus-32 were each identified once in six samples. Other radionuclides for which analysis in alfalfa was made were not identified.

H. Soil

Since all fallout components will at least in part be deposited on the soil, characterization of soil samples is difficult. The general criteria for source and time of release and nature of release apply. Cesium-137 is held in the soil rather tightly in comparison to strontium-90. The residual soil contamination in the Marshall Islands two years after weapons testing is shown in Table 8.(37)

Table 8. Soil nuclide composition

Depth 0-1 inch

Nuclide	Percentage of Total
Cesium-137	$< 1\%$
Strontium-90	5
Ruthenium-106	10
Total Rare Earths	84

Possible induced activities in soils have been determined by a number of investigators.(18, 19, 38) The more common include sodium-24, phosphorus-32, potassium-42, calcium-45, iron-55, iron-59, and manganese-56.

V. SUMMARY

The nature of contamination of an environmental sample must be known for efficient quantitative gamma spectral analysis of the sample. Any knowledge of the source of contamination, nature of release, and time since the contamination will be helpful since these factors will modify the expected contamination. A knowledge of the food chain discrimination and other environmental factors will help eliminate unlikely or impossible radionuclides from consideration in qualitative and quantitative gamma spectroscopy.

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Fission Product Age Determination

I. INTRODUCTION

Radionuclide analysis of environmental media can be greatly facilitated by a knowledge of the sample. One parameter which is of particular value in the analysis of fission products in air is their apparent age. This provides an initial screening device for qualitative analysis. In other words, if the age of the fission product is determined, then all radionuclides with a half-life less than one-seventh of the apparent age may be eliminated from initial consideration. These nuclides will have undergone seven half-lives of decay and will be about 99.5% decayed out.

The following discussion presents a procedure by which the approximate fission product age may be determined for relatively fresh fission product material.

II. THE WAY-WIGNER FORMULA

It has been determined experimentally that fission products display a more or less constant relationship between age and apparent beta activity. This relationship states that:

$$At^{1.2} = \text{constant}^{(1)} \qquad 1)$$

where

A = Beta activity

t = time after fission product formation

It is axiomatic that:

$$A_1 t_1^{1.2} = A_2 t_2^{1.2} \qquad 2)$$

where: t_1 and t_2 are elapsed times since fission occurred and,

A_1 = Beta activity at time t_1

A_2 = Beta activity at time t_2

Using the above relationship (equation 2), the apparent age of fission products in an air sample may be calculated in terms of the activities measured at two different times and the time interval (Δt) between the two counts. Solving the equation for t_1, and remembering that $\Delta t = t_2 - t_1$, the following relationship is derived.

$$t_1 = \frac{\Delta t_1}{\left\{\left(\frac{A_1}{A_2}\right)^{0.835} - 1\right\}} \qquad 3)$$

where:

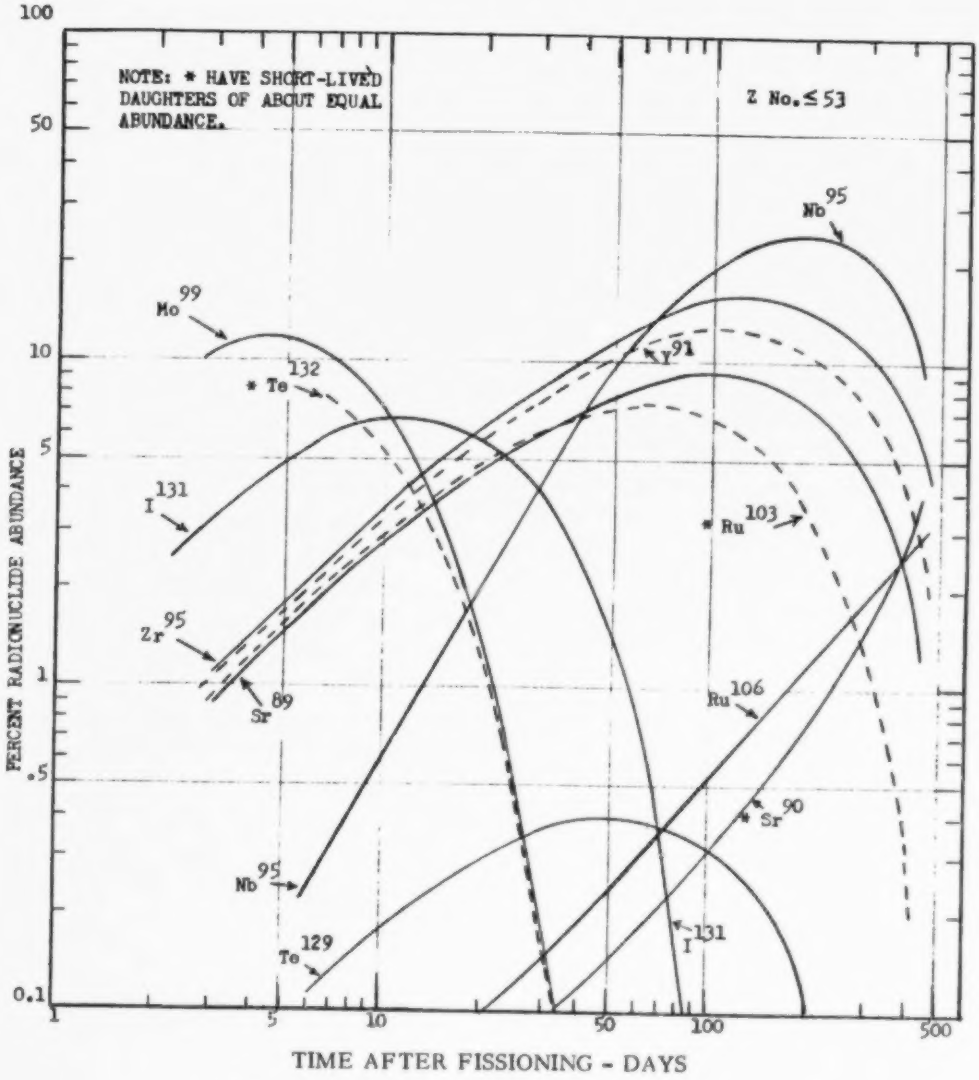
t_1 = apparent age at time of first counting

Δt_1 = time elapsed between first and second counting

A_1 = Beta activity at first counting

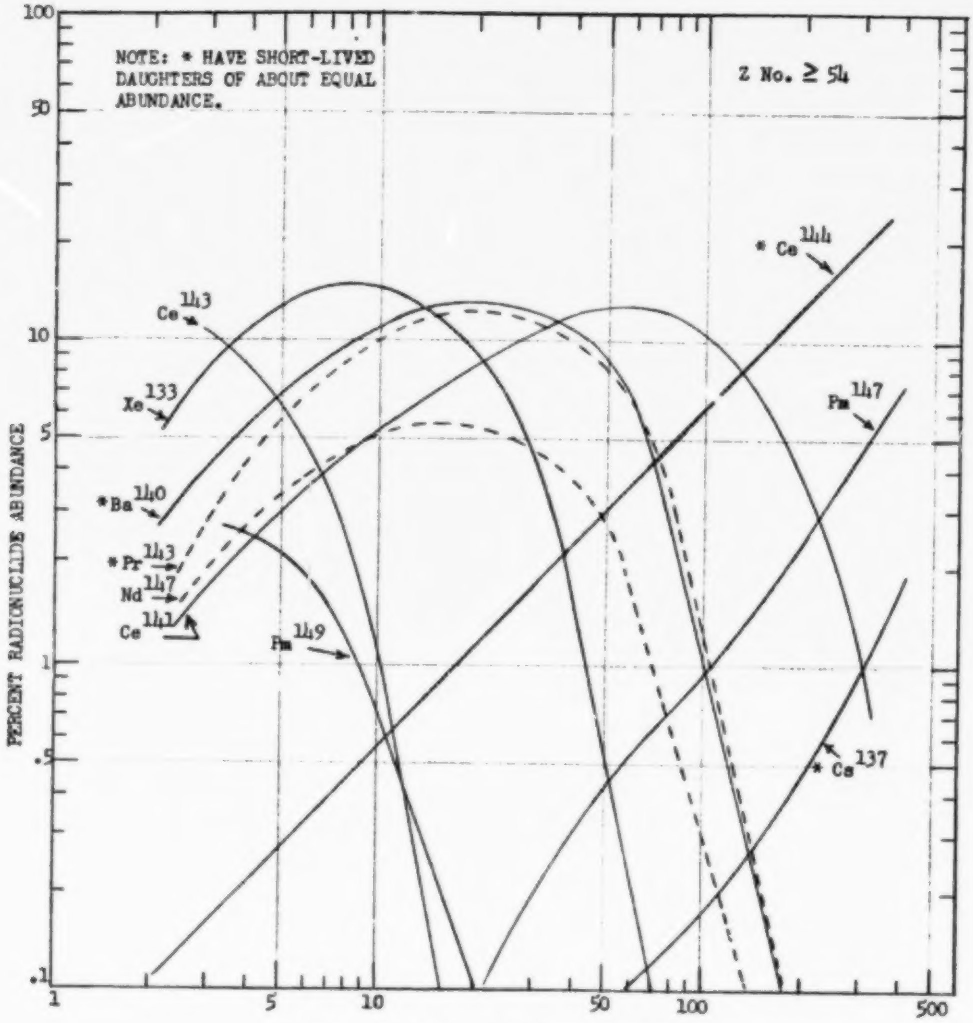
A_2 = Beta activity at second counting

This equation gives the apparent age of the fission products at the time of the first count.



Relative Abundances of Fission Product Nuclides
Up to 500 Days After Fissioning

Figure 2



TIME AFTER FISSIONING - DAYS

Relative Abundances of Fission Product Nuclides
Up to 500 Days After Fissioning

Figure 2a

III. PERCENT RADIONUCLIDE ABUNDANCE

Once the average age of the fission products is known, one may proceed to a percent radionuclide abundance chart such as is shown in Figures 2 and 2a. From such a chart, those nuclides still remaining at the time of sampling can be determined.

IV. SUMMARY

The quantitative gamma analysis of air samples can be greatly facilitated by a knowledge of the apparent age of the fission products. The Way-Wigner Formula may be applied to determine the apparent age. Once the age of the fission products is known, those nuclides of comparatively short half-lives may be eliminated from initial consideration.

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Chemical Simplification of Complex Spectra

I. INTRODUCTION

It is often necessary to perform chemical separations on complex samples to simplify the resulting spectra. The degree of simplification of spectra needed is variable in each case and is often a matter of judgment. A sample can be simplified to the point that gamma counting can be performed on gross gamma counters or single channel analyzers. In other cases samples may be only partially simplified and subsequent counting of "simplified" fractions may still require multichannel analyzers.

In most cases, chemical separations are qualitative in nature and are used for identification rather than quantitative analysis. However, by adding standardized carriers and performing chemical yields or duplicating conditions from one sample to another, it is possible to do quantitative analysis with a minimum number of steps.

II. SIMPLIFICATION OF COMPLEX SAMPLES

A. Milk

Since milk has been one of the main contributors of iodine-131 to the human diet, a great deal of effort has been directed toward developing simple, rapid techniques for determining iodine-131 in milk, involving gamma counting of the separated iodine. Two of these techniques are outlined below:

1. PRECIPITATION OF PROTEINS(1)

The formalin which is routinely added to milk as a preservative causes the iodine-131 in the milk to become chemically bound to the proteins. The proteins can be precipitated from the milk by adding trichloroacetic acid. The proteins are then recovered by filtration, transferred to a plastic bottle and counted on a gross gamma counter or a single-channel analyzer.

2. ABSORPTION BY ANION EXCHANGE RESIN RESIN(2,6,7)

It is known that in fresh milk, about 90-95% of the iodine-131 is present as inorganic iodide ion. The inorganic iodide-131 can be removed from milk by pouring the milk through an ion exchange column containing an anion exchange resin in the chloride form. The resin can then be transferred to a plastic bottle and counted on a gross gamma counter or a single-channel analyzer. It is common practice to calibrate the gamma counter by adding calibrated iodine-131 to distilled water and obtaining counting efficiencies at various volumes. A calibration curve of efficiency versus volume of distilled water is then constructed, and the efficiency for iodine-131 is determined from the volume of resin or protein. Again, the density of resin and protein may be slightly different than water, but for an approximate method the difference is not restrictive.

B. Water

Certain water samples (rainfall containing fresh fallout(3), surface water near nuclear installations(8), and reactor cooling water(4, 8)) can exhibit complicated gamma spectra. If the radionuclide composition of the sample is known or can be determined, then it is possible to set up and solve a set of simultaneous equations for these radionuclides. The Cooperative Studies Unit of the Radiological Health Research Activities of the Robert A. Taft Sanitary Engineering Center is currently using a series of nine equations for solving the concentrations of nine radionuclides. These solutions are performed on a computer. If such solutions are not practicable, then the samples often have to be simplified by chemical analysis.

1. QUALITATIVE ANALYSIS SEPARATIONS[4]

A knowledge of qualitative chemical analysis is necessary in order to carry out a logical, orderly separation of the radionuclides into elemental chemical groups. A brief outline of the classical qualitative analysis group separations which can be found in any of the standard textbooks on qualitative analysis is summarized in Table 1.

The application of a qualitative analysis scheme, such as the one presented in Table 1, to the simplification of gamma spectra necessitates considering the various group separation with respect to their radionuclide content. A correct interpretation of a "simplified fraction" will depend not only on associating specific radionuclides with the appropriate group separation, but also on the ability of the analyst to identify unknown peaks which may be due to either unsuspected radionuclides or incomplete group separations.

A separation scheme which has proved to be effective in separating radionuclides in reactor cooling water is given in Table 2.[4] This method is based on the standard qualitative analysis separation as listed above.

The separated groups can be counted on a gamma spectrometer and the resulting spectra are much simpler than the original sample.

2. BASIC SULFIDE PRECIPITATION[5]

A method which was developed for determining gross radioactivity in sea water also has applications to simplifying gamma spectra. This technique eliminates several of the classical qualitative analysis steps and attempts to concentrate most of the radionuclides of concern into three groups, basic sulfide, alkaline earth, and soluble groups. A brief outline of the scheme is given in Table 3.

3. SOLVENT EXTRACTION OF IODINE-131[6]

It is sometimes useful to be able to isolate pure iodine-131 from a water sample. This

Table 1. Qualitative analysis group separations

Group	Group reagent	Cations precipitated
Silver	Dilute HCl	Ag, Pb, Hg(I)
Acid Sulfide	H_2S in Acid Solution	Pb, Bi, Cu, Cd, As, Sb, Sn, Hg(II)
Basic Sulfide	$(NH_4)_2S$ or H_2S in Ammoniacial Solution	Sulfides of Zn, Ni, Co, Mn, Fe(II) Hydroxides of Al, Cr, Fe(III)
Alkaline Earth	$(NH_4)_2CO_3$ in Ammoniacial Solution	Ca, Sr, Ba
Soluble	None	Solution of Mg, Na, K

Table 2.4 Chemical separation scheme

1) Add an aliquot of the radioactive sample to a 50 milliliter centrifuge tube. Aerate to remove radioactive gases. Acidify with HNO_3 and add Cl, Br, I, Cu, As, Fe, Y, Co, Mn, Zn, Ba, Sr, Ca, Na and K carriers. Precipitate Cl, Br, and I with slight excess of Ag. Heat to coagulate the precipitate. Centrifuge.

- 2) Precipitate: AgCl, AgBr, AgI, Wash with water, filter, mount and count. Designate as the Silver Group.
- 3) Supernate: Adjust acidity to 0.5 N HCl. Saturate with H_2S. Centrifuge.
  - 4) Precipitate: $CuS + As_2S_3$. Wash with water, filter, mount and count. Designate as the Acid Sulfide Group.
  - 5) Supernate: Transfer to beaker. Add 1 ml. CONC. HCl. Boil until solution is clear. Oxidize Fe^{++} to Fe^{+++} by Boiling with HNO_3. Cool solution and make 1:1 with CONC. HCl. Neutralize with CONC. NH_4OH to precipitate hydroxide. Centrifuge.
    - 6) Precipitate: Fe + Y hydroxides. Wash with water, filter, mount and count. Designate as the Hydroxide Group.
    - 7) Supernate: Saturate with H_2S. Centrifuge.
      - 8) Precipitate: CoS, MnS, ZnS. Wash with water, filter, mount, and count. Designate as the Basic Sulfide Group.
      - 9) Supernate: Acidify solution with HCl. Boil to remove H_2S. Add $(NH_4)_2H(PO_4)$ and an excess of NH_4OH. Digest. Centrifuge.
        - 10) Precipitate: Ca, Ba, and Sr phosphates. Wash with water containing a few drops of NH_4OH. Filter, mount and count. Designate as the Alkaline Earth Group.
        - 11) Supernate: Na + soluble ions. Transfer aliquot to culture tube for counting. Designate as the Soluble Group.

Table 3.^{5} Basic sulfide precipitation

1) Add La, Fe(III), Co(II), Zn, Ni, Cs, and Sr carriers to 1 liter of sea water, acidify, boil, and make basic with NH OH. Precipitate sulfides with thioacetamide or H_2S. (Tracers added in development were Ce^{144}-Pr^{144}, Cr^{51}, Ru^{106}-Rh^{106}, Zn^{65}, Co^{60}, Zr^{95}-Nb^{95}, Sr^{85}, Cs^{137})

2) Precipitate: La, Fe, Co, Zn, Ni Ce^{144}-Pr^{144}, Cr^{51}, Ru^{106}-Rh^{106}, Zn^{65}, Co^{60}, Zr^{95}-Nb^{95} either as sulfides or hydroxides: dissolve precipitate in HCl-HNO_3 and gamma count. (Sulfide ppt. may be beta counted, or further chemical separations may be performed).

3) Supernate: Acidify with HCl, boil, neutralize, add 8-hydroxyquinoline to remove Mg. filter, make supernate basic with NaOH, add Na_2CO_3.

4) Precipitate: Sr, Ba, Ca, and Ra as carbonates, filter, dissolve carbonate in 30 ml. CON HNO_3, repeat, dissolve nitrate in water, reprecipitate carbonates, beta count.

5) Supernate: Cs^+, K^+, and other soluble ions in solution. Supernate may be gamma counted directly or cesium ammonium phosphomolybdate for later beta or gamma counting.

may be rather easily accomplished by a carbon tetrachloride solvent extraction procedure which is selective for iodine.

a. To the original sample add iodide carrier, NaOH, and NaOCl to oxidize carrier and radioiodine. Add 6 N HCl and $NaHSO_3$ to reduce carrier and radioiodine to iodide. (This assures complete interchange.)

b. Add $NaNO_2$ and extract free iodine into CCl_4.

c. Back extract iodine from CCl_4 with $NaHSO_3$.

d. The NaHSO portion can then be gamma counted on a gross counter or single channel analyzer.

C. Air filters

During periods when fresh fission products are present, air filter samples may exhibit complicated gamma spectra. In some cases it may not be possible to obtain the desired information by spectrum stripping and it is necessary to resort to chemical separation procedures for definite identification and quantitative assay of radionuclides present. Outlined below is a procedure for the chemical simplification of one type of filter paper.

1. Glass Fiber Paper - Used in National Air Sampling Network - Preparation of Glass Fiber Paper by Caustic Fusion

While this method is not one which is being performed on a routine basis, it is noteworthy as a good example or preparing glass fiber filters for analysis.

a. Place filter paper in nickel crucible with iodine, ruthenium, barium, zirconium, niobium, tungsten, and cesium carriers. Fuse with NaOH-Na_2CO_3. Break up fusion melt with hot water and centrifuge.

b. The supernate contains I^{131}, $Ru^{103, 106}$, W^{185}, Cs^{137}, and a small amount of Ba^{140}-La^{140}. The ruthenium may be precipitated by alcohol reduction, the iodine extracted by CCl_4, tungsten oxide precipitated by heating, and cesium precipitated as phosphomolybdate.

c. The precipitate from the fusion contains Sr^{89}, Sr^{90}-Y^{90}, Ba^{140}-La^{140}, Zr^{95}, the rare earths, and small amounts of $Ru^{103, 106}$. This precipitate may be dissolved with HCl, and all of the above mentioned activities reprecipitated with NH_4OH and Na_2CO_3. A fuming nitric acid separation gives a precipitate containing Sr, Ba, and Nb and a supernate containing Zr, the rare earths, and small amounts of $Ru^{103, 106}$.

III. SUMMARY

Again, it should be emphasized that the separated fractions for any simplification process present unique calibration problems. The separated fractions may still be so complex that further separation is necessary. Usually, though, the standard analytical techniques such as spectrum stripping or solution of simultaneous equations can be applied to each fraction to yield the desired results.

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Compton Continuum Subtraction Method

(Quantitative Analysis)

I. INTRODUCTION

Since several radionuclides may have similar gamma photon energies, and because of the contribution to lower energy channels from Compton scattering, the gamma spectra representing a combination of several radionuclides, as is often encountered in environmental samples, may be very complex, and thus quite difficult to interpret.

The purpose of this lecture is to present an approximate, graphical procedure for the quantitative analysis of complex environmental gamma spectra.

II. COMPTON CONTINUUM SUBTRACTION METHOD

A. General Considerations

The objective of this method is to eliminate by approximation the contribution to a given photopeak by interfering radionuclides. By summing up the count rates for several selected channels within the characteristic photopeak, and then approximating the portion of these count rates contributed by interfering radionuclides, the actual activity due to the single radionuclide can be estimated. If the appropriate counting efficiency is then applied, the rate of disintegration can be determined.

B. Choice of photopeaks

Since the characteristic photopeaks are the distinguishing features between gamma spectra, the efficiencies and therefore the rates of disintegration are usually determined for only selected channels within the photopeaks. Many radionuclides display more than one distinguishing photopeak however, and a choice must be made as to which is the most indicative. See Figure I.

There are several considerations which may influence the choice of photopeak.

1. It is essential to select peaks which do not overlap with peaks of other radionuclides. The graphical procedure cannot discern between such overlapping peaks.

2. It is desirable to select peaks exhibiting a high total counting efficiency. This encompasses two factors:

   a. Gamma abundance.

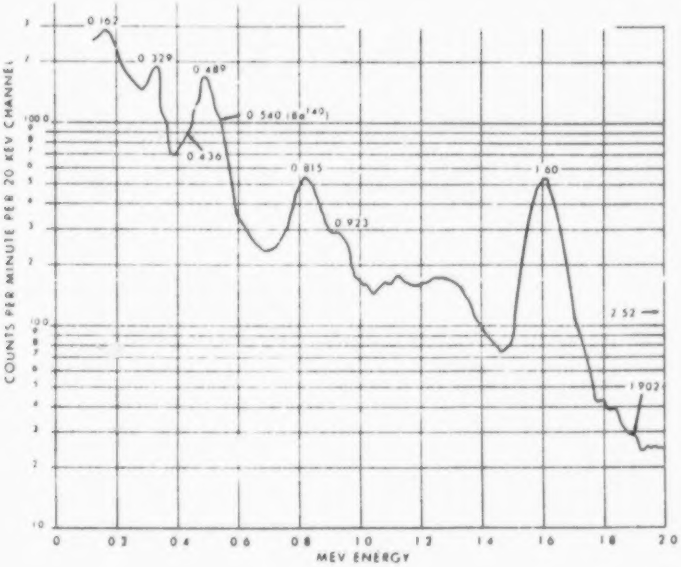
   b. Gamma energy (low energy photons will have a higher intrinsic efficiency resulting in a higher photopeak to Compton ratio).

3. Low energy peaks are less influenced by gain shift.

4. High energy peaks are subject to less Compton interference.

5. High energy peaks yield better resolution characteristics by definition.

It is important to realize that the above criteria can be used only as a rough guide line. Each spectrum encountered must be reviewed individually as to its own characteristics before a choice of photopeaks is made.



STANDARD SPECTRUM FOR Ba^{140} - La^{140} SHOWING MULTI-PHOTOPEAK SPECTRUM

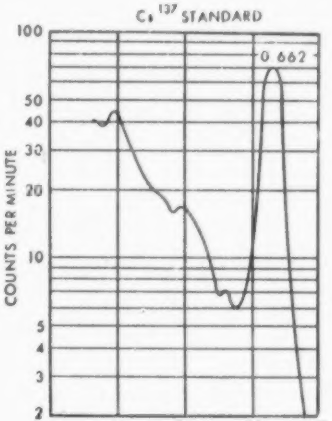
Figure 1

C. Choice of channels

Once a photopeak is selected it becomes necessary to determine how many channels within the photopeak should be used for analysis. This choice becomes especially critical when considered in relation to gain shift. The safest and most reproducible procedure is to use the peak channel plus an equal number of channels on each side such that, for a gain shift of one channel, all the selected channels will still fall within the photopeak. Because of the nearly normal shaped curve, the counts lost during the gain shift will then be approximately equal to the counts gained.

As seen in Example 1, using only the peak channel, or using the total photopeak channels produces a larger gain shift error than that resulting from the use of selected channels within the photopeak.

Example 1 - Effect of channel selection on gain shift error



CESIUM-137 PHOTOPEAK

Figure 2

DATA FOR FIGURE 2

Channel No.	Count Rate
26	7.11
27	6.03
28	6.14
29	7.47
30	14.56
31	34.35
32	63.05
33	67.05
34	50.14
35	23.51
36	8.32

- Assuming a 1 channel gain shift

a. Using peak channel only (ch 33)

Count without gain shift - 67.05
Count with 1 ch. gain shift - 63.05

Difference - 4.00

$$\% \text{ Error} = \frac{4}{67.05} \times 100 = 5.96\%$$

b. Using photopeak channels (30-36)

Count without gain shift - 260.98
Count with 1 ch. gain shift - 260.13

Difference .85

$$\% \text{ Error} = \frac{.85}{260.98} = 0.33\%$$

c. Using total photopeak area (ch. 27-36)

Count without gain shift - 280.62
Count with 1 ch. gain shift - 279.41

Difference 1.21

$$\% \text{ Error} = \frac{1.21}{280.62} = 0.43\%$$

D. Procedure

1. Approximating the general slope of the entire spectral curve, draw a smooth line estimating the base of the peak being considered.

2. Sum up the observed count rates for the selected channels and subtract from this total the sum of the count rates indicated by the base line (these are the counts contributed by scattering from other radionuclides) for the same channels.

3. Using the appropriate efficiency factor for the selected channels, calculate the disintegration rate. It should be remembered that the counting efficiency must be determined by the same approximate method (i.e., subtracting out Compton contribution) using single radionuclide standards. This procedure is illustrated in Example 2.

Example 2

The spectral data shown in Figure 3 was obtained from a 3.5 liter water sample. The 0.66 Mev Cs^{137} photopeak will be considered. The shaded area in this Figure indicates the count rates resulting from scatter that must be subtracted from the contribution from Cs^{137}. Assume a total counting efficiency of 4%.

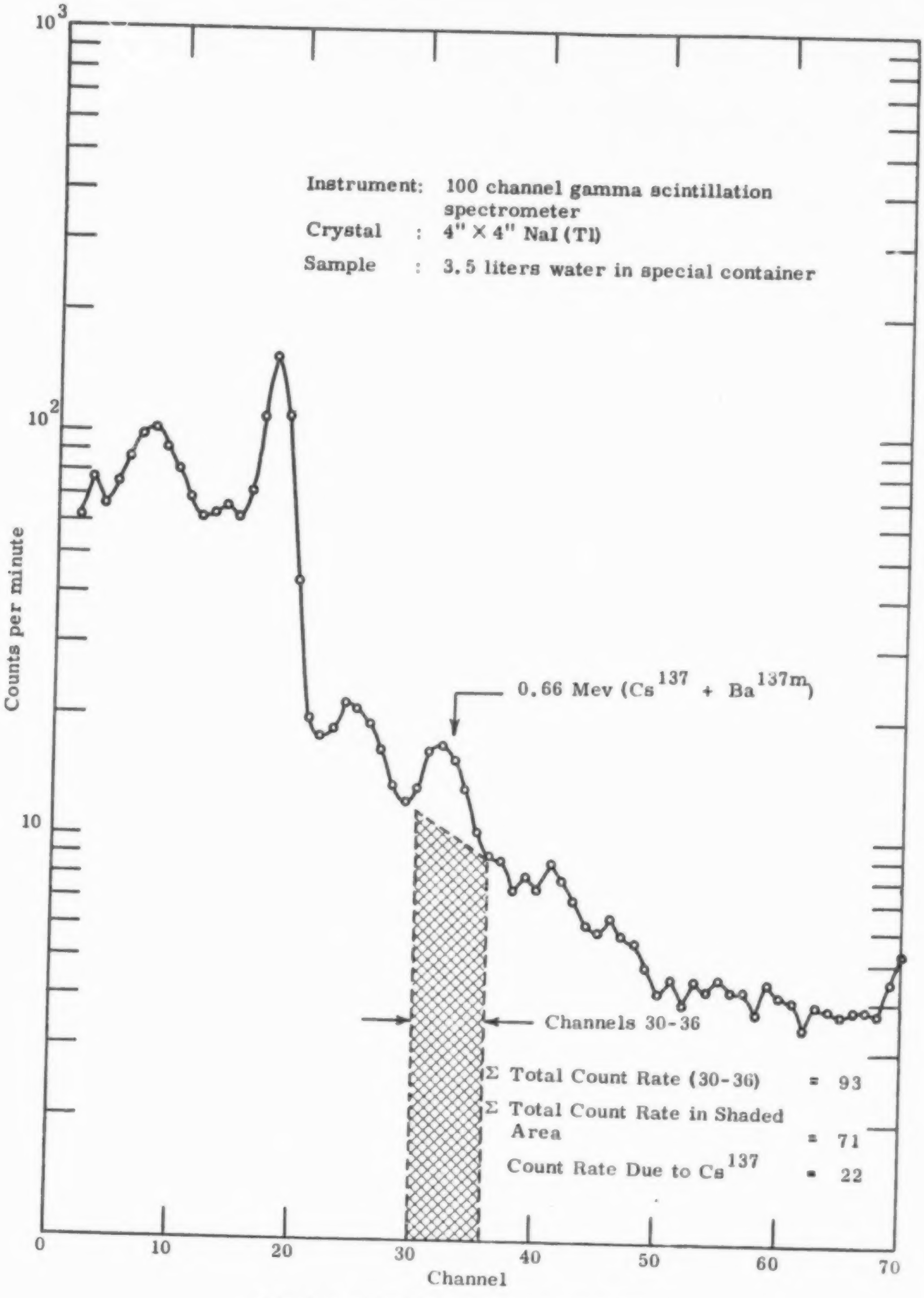
The calculations are as follows:

Peak count rates, channels 30-36 = 93
Count rates from scattering (shaded area) = 71

Total count rate due to Cs^{137} = 22

$$\text{Disintegration rate} = \frac{\text{Observed Count Rate}}{\text{Efficiency}}$$

$$\text{Disintegration rate} = \frac{22}{0.04} = 550 \text{ dpm}$$



GAMMA SPECTRAL DATA FOR WATER

Figure 3

III. SUMMARY

This method provides a simple procedure for the quantitative analysis of environmental gamma spectra. It should be kept in mind however, that it is only an approximate, graphical determination. For more precise analysis, other, more refined methods must be employed.

Spectrum Stripping Method

(Qualitative and Quantitative Analysis)

I. INTRODUCTION

A. Statement of Problem

In most instances the analysis of a gamma spectrum involves both qualitative identification and quantitative evaluation of the various component nuclides. The complexity of the spectrum to be analyzed is due to many factors, among them being:

1. Compton interference from higher energy gamma photons to lower energy photopeaks.

2. The presence of several radionuclides, each of which may emit several different energy gamma photons, thus producing many photopeaks.

3. The existence of secondary peaks; e.g., escape, annihilation and sum.

4. The presence of nuclides that have gamma-emitting daughter products "growing in" due to series decay.

5. Wide variations in the relative activity of each nuclide.

6. Various detection efficiencies for different energy photons.

7. The non-uniqueness of gamma photon energies which results in hidden or overlapping peaks.

One method that may be used to perform both qualitative and quantitative analysis is "spectrum stripping". It may be used independently or in conjunction with other methods, depending on the complexity of the spectrum.

B. Principles Utilized

When a mixture of gamma rays from two or more different radionuclides is being observed, the resultant composite spectrum will be the channel-by-channel summation of the spectra of the individual radionuclides in the mixture. Therefore, if the spectrum of a single radionuclide can be obtained, it can be subtracted channel-by-channel from the composite spectrum, resulting in a spectrum containing one less nuclide than the original. This process is termed "spectrum stripping". Each individual spectrum which is subtracted is obtained by utilizing a previously counted "standard" of the radionuclide and multiplying it, channel-by-channel, by a spectrum multiplier--to be discussed later. The use of "standards" is made possible by the assumption that the relative shape of a spectrum for a single nuclide is independent of its activity. Difficulties associated with this process will be discussed subsequently.

C. Usefulness of Spectrum Stripping

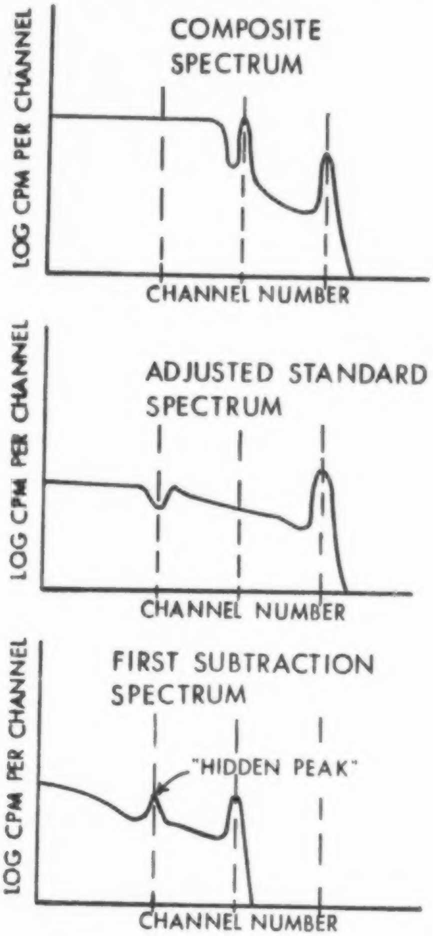
Spectrum stripping can be a valuable tool in qualitative identification and quantitative evaluation of a gamma spectrum.

1. QUALITATIVE IDENTIFICATION

By stripping the contribution of known nuclides from a composite spectrum, nuclides previously hidden may be uncovered, see Figure 1. The uncovered peaks will have been previously hidden due to the presence of the nuclide spectrum stripped out.

2. QUANTITATIVE EVALUATION

The only region of a composite spectrum where the peak height (cpm per channel) or photopeak area (cpm) of a single peak is proportional to the activity of the nuclide which caused the peak, is the highest energy peak of the spectrum. The lower energy photopeaks, will, in part, be due to Compton interference from all higher energy photopeaks, while the highest energy peak is normally free from any interference. The exception to this is when the highest energy peak is in actuality a combined peak composed of similar energy gamma rays from two independent nuclides.



SUBTRACTION OF SINGLE SPECTRUM
FROM COMPOSITE SPECTRUM
Figure 1

The activity of the nuclide causing the highest energy peak can therefore be obtained directly. If the spectrum associated with this peak is subtracted from the composite spectrum, yielding a "first subtraction spectrum", the next highest energy peak will become interference-free and the activity may be determined. This process may be continued until all peaks are cleared.

II. METHODS OF SPECTRUM STRIPPING

A. Procedure

1. STEPS

The suggested steps in a spectrum stripping procedure are as follows:

a. Determine the presence of as many nuclides in the sample as possible, especially those with higher energies. This identification can be made by using peak energy, half-life, knowledge of sample, etc.

b. Beginning with the highest energy peak nuclide, determine the spectrum multiplier (S. M.) to be applied to the counting data of the standard spectrum of that nuclide. The spectrum multiplier (S. M.) is equal to the ratio of the cpm in the photopeak area of the nuclide in the composite spectrum to the cpm in the photopeak area of the standard spectrum for that nuclide.

$$\text{S. M.} = \frac{\text{cpm photopeak area (Sample)}}{\text{cpm photopeak area (Standard)}}$$

The S. M. can also be obtained by determining the peak channel cpm per channel ratio; however, this value is not as reliable as the S. M. obtained by using the photopeak area ratios.

The S. M. can be obtained directly from the composite spectrum when the standard spectrum peak has either its peak channel cpm or photopeak area normalized.

c. Multiply the count rate of each channel of the standard spectrum by the S. M., and then subtract each channel cpm value of the adjusted standard spectrum from the sample composite spectrum.

d. Determine the activity of the nuclide "stripped" by dividing either the peak channel cpm, photopeak area cpm, or total cpm by the appropriate counting efficiency.

e. Repeat steps b, c, and d for the highest energy peak nuclide in the "first subtraction spectrum". Continue until only one nuclide from the original spectrum remains, then apply step d to this nuclide.

2. RADIONUCLIDE STANDARDS

A "standard" spectrum for each radionuclide suspected of appearing in the composite spectrum is a requirement of spectrum stripping. The spectral shape for a radionuclide is dependent on the energy of the gamma ray(s) emitted, the relative abundance of the gamma rays when more than one is emitted, and the physical environment where the counting is made. Some factors involved in the physical environment are the size and shape of the crystal and sample container, the thickness of the crystal canning, and the presence of external shielding. Since there are so many factors affecting the spectral shape, it is imperative that the standard spectrum and sample spectrum be obtained under as nearly identical physical environment conditions (counting arrangement) as possible. This implies that standard spectra obtained on one spectrometer cannot be used for spectrum stripping of a sample counted on another spectrometer.

B. Total Spectrum Method (Manual)

The procedure described previously can be done by the use of a slide rule and adding machine or a calculator. Although hand methods are tedious and time consuming, the total time and cost for sample analysis is less than that required when radiochemical procedures for separating sample components are utilized.

C. Total Spectrum Method (Graphical)

An approximate spectrum stripping method, which eliminates the need for spectrum multipliers, can be accomplished by cutting the standard spectra along the ordinate values (log cpm per channel), placing this template on the spectra to be stripped, and making graphical channel-by-channel subtractions. As the shape of the log plot is linearly constant, any arbitrary baseline may be chosen. Thus, by simply overlapping an interference free peak with the template, which automatically determines the S. M., the Compton portion of the template may be subtracted from the composite curve.

D. Total Spectrum Method (Commercial)

Commercial spectrum stripping instruments are available which, when used in conjunction with the gamma spectrometer memory core, perform spectrum stripping semi-automatically.

E. Partial Spectrum Method (Manual)

In some instances it is not necessary to completely strip a nuclide from a spectra being analyzed, stripping a portion or a few selected channels of interest may be satisfactory. This can only be done when all nuclides present in the sample are known and a quantitative evaluation is being made; i.e., on a routine basis for milk if simultaneous equations are not available.

III. DIFFICULTIES ASSOCIATED WITH SPECTRUM STRIPPING

There are several difficulties associated with the spectrum stripping method which could limit its precision and usefulness. Some of these difficulties will be discussed below.

A. Subtraction Errors

1. EFFECT

There are errors associated with every spectrum which may accumulate to a serious degree after several nuclides have been stripped. Therefore, the quantitative evaluation of lower energy gamma emitters is less accurate than the higher energy emitters because of the error build-up in the lower energy channels.

2. CAUSE

The errors will result mainly from the fact that the shape of an individual nuclide spectrum in a composite spectrum is not an exact replication of the shape of its "standard" spectrum. As a result of this inability to obtain perfectly reproducible spectrum shapes, the spectrum multiplier utilized in spectrum stripping will be somewhat in error.

Gain variation is the major factor causing non-reproducibility of spectrum shapes (peak shifts). Gain variations may be caused by changes in line voltages, gain drift with time, or imperfect instrument calibration between "standards" counting and sample counting.

3. ALLEVIATION

Frequent calibration of the complete spectrometer system, by using standard radionuclide sources or an electronic pulser, is essential to the detection and correction of peak shifts due to gain variations.

B. Coincidence

In samples containing two or more radionuclides emitting different energy gamma rays, there is a small probability that two or more of these different energy gamma rays from different nuclides may be detected in coincidence, thus causing "sum" energy peaks to occur. Sum peaks simply denote peaks that have energies corresponding to the summation of the energies of the individual components. Sum peaks will also have an associated sum spectra of energy due to the detection of various combinations of individual peak energies and Compton gamma energies, or combinations of Compton gamma energies, all of which may occur in coincidence.

Since environmental samples may contain several gamma emitting nuclides, the possibility of the occurrence of sum peaks and their associated sum spectra exists. Therefore, systematic stripping of identified nuclides will not account for the sum peaks and their sum spectra; however, the sum spectrum is usually much smaller, relatively, than the rest of the spectrum, and other sources of error are usually larger than those due to neglecting the sum spectra, so it is neglected.

The occurrence of sum peaks for a single radionuclide are of no consequence because they will be indicated on the "standard" spectrum and subsequently subtracted in the process of stripping that nuclide.

C. Overlapping Peaks

Instances may arise in sample analysis where peaks from two or more gamma rays of similar energies overlap. In other words, instead of obtaining discrete peaks spaced closely together, one wide energy span peak may occur containing the discrete peaks. Overlap creates problems in both qualitative identification and quantitative evaluation.

1. APPARENT ACTIVITY METHOD

When the nuclides in the overlapping peak have no other distinguishable peaks in their spectrum the activity of peak can be reported as "apparent activity" of the suspected nuclides (assuming equal gamma abundances and gamma detection efficiencies), and the contribution of this peak stripped if the shape of all the suspected nuclides is approximately the same; i.e., Zr^{95}-Nb^{95}.

2. TRIAL-AND-ERROR METHOD

When one of the nuclides in the photopeak has another distinguishable peak associated with it, a "trial-and-error" stripping procedure can be used. In this process, "trial-and-error" amounts of the suspected nuclide are stripped until the associated peak disappears from the composite spectrum, thus indicating that all of the contribution of the suspected nuclide has been stripped from the overlapping peak.

V. SUMMARY

Spectrum stripping can be a very useful tool in gamma spectral analysis even though it is tedious and time-consuming. Spectrum stripping would probably not be done on a normal, routine basis; however, it provides a good "spot check" for sample contamination by unidentified nuclides, and for infrequent checking of matrix equations.

If the steps in spectrum stripping methods are rigidly followed, even though there are inherent process errors, meaningful results can be obtained.

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GPO 864-519

Simultaneous Equation Method

(Quantitative Analysis)

I. INTRODUCTION

Many types of environmental samples, which must be analyzed routinely, have a fairly constant radionuclide content which is limited to a specific minimal number of nuclides. Examples are milk contaminated with 10 day or older fallout, 30 day or older atmospheric fallout particulates, or effluents from normal reactor operations if short-lived nuclides are decayed out.

A convenient method for the quantitative analysis of this type of sample is the "simultaneous equation method." This method entails a mathematical approach for eliminating Compton interference from photopeaks. The only requirements for application of this method are:

1) That all nuclides present in the sample are identified.

2) That a different photopeak be present for each nuclide in the sample. This does not exclude the possibility of many overlapping peaks.

II. DEVELOPMENT OF METHOD

A. Interference Factors

The principle of "constant spectral shape" states that the ratio of any portion of a spectrum to another portion is constant and independent of the activity. A useful ratio is that of the photopeak channel(s) of a different nuclide to the photopeak channel(s) of the nuclide whose spectrum is being considered. This ratio, termed an interference factor, is obtained from a standard spectrum of the single nuclide. The significance of this terminology will become apparent shortly.

First consider the spectrum of the nuclide Z (see Figure 1). It is possible to express the count rate of any channel in terms of any other channel. If channel 10 is taken as the reference peak, the count rate in channel 20 can be written as:

$$CR_{20} = f_{(10-20)} CR_{10}$$

where

CR_{20} = count rate in channel 20

CR_{10} = count rate in channel 10

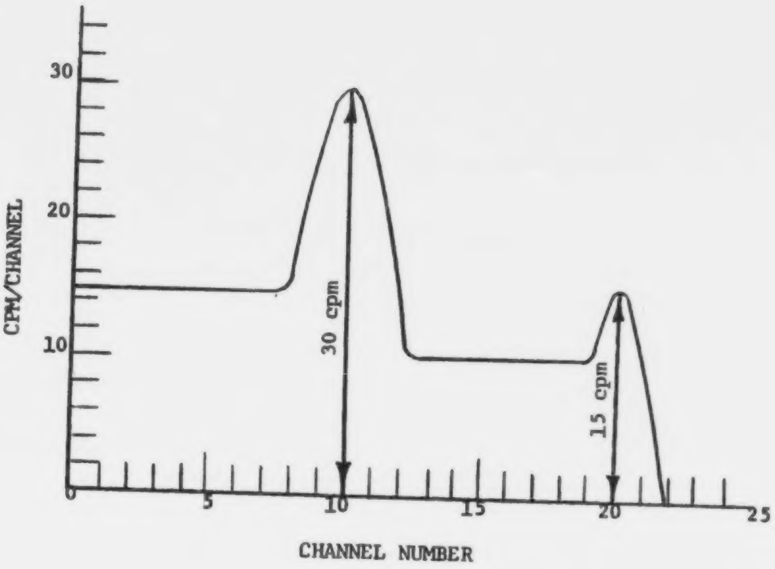
$f_{(10-20)}$ = interference factor

The interference factor is calculated by:

$$f_{10-20} = \frac{CR_{20}}{CR_{10}} = \frac{15}{30} = 0.5$$

thus if count rate in channel 10 of another spectrum of nuclide Z is 50 counts per minute, the count rate in channel 20 becomes 25 cpm.

$$CR_{20} = 0.5 \times 50 = 25 \text{ cpm}$$



SPECTRUM OF NUCLIDE Z

Figure 1

B. Simultaneous Equations

Next the spectrum of a sample containing two radionuclides, X + Y is considered. Figure 2 shows the individual spectra of X and Y and the composite spectrum.

The net count rate of the composite spectrum for the photopeak channel of nuclide X (N_x) can be written as:

$$N_x = X_x + X_y \qquad (1)$$

where

X_x = count rate contributed by nuclide X to the photopeak channel of X

X_y = count rate contributed by nuclide Y to the photopeak channel of X

However, X_y may also be expressed in terms of Y_y, the count rate contributed by nuclide Y to the photopeak channel of Y.

$$X_y = f_{yx} Y_y \qquad (2)$$

where

f_{yx} = interference factor which expresses the fractional contribution of the photopeak channel of nuclide Y to the photopeak channel of nuclide X.

Combining equations (1) and (2), we have:

$$N_x = X_x + f_{yx} Y_y$$

A similar development can be done for the net count rate in the photopeak channel of nuclide Y (N_y), resulting in the equation:

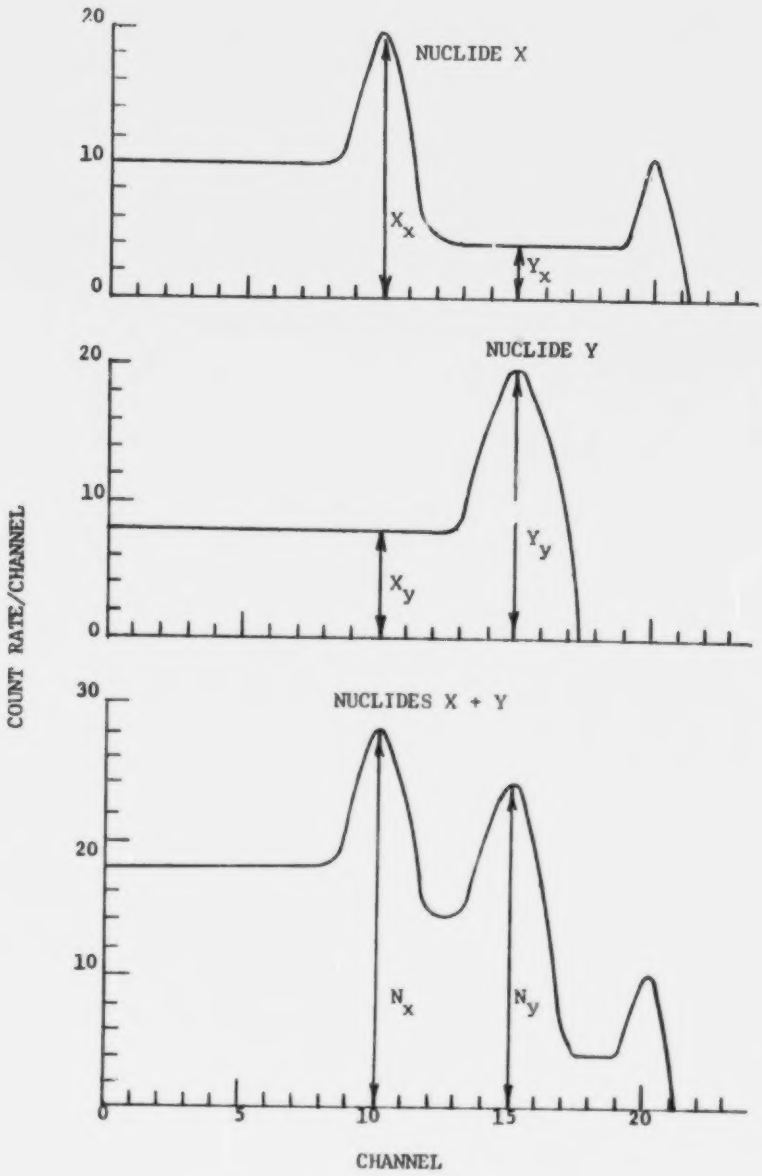
$$N_y = Y_y + f_{xy} X_x$$

C. Solution of Simultaneous Equations

The two equations:

$$N_x = X_x + f_{yx} Y_y$$

$$N_y = Y_y + f_{xy} X_x$$



SPECTRUM OF COMPOSITE SAMPLE

Figure 2

where

C_1, C_2, C_3, C_4 are dimensionless coefficients

D. Calculation of Activity Concentration

The activity concentrations (pc/1) for each nuclide in a composite sample are calculated by the equation:

$$\text{Nuclide X pc/1} = \frac{X_x}{(\text{eff})(\text{Vol})(2.22)}$$

$$\text{Nuclide Y pc/1} = \frac{Y_y}{(\text{eff})(\text{Vol})(2.22)}$$

are simultaneous linear independent equations containing two unknowns, X_x and Y_y. The net count rates (N_x, N_y) of the composite sample are obtained from the composite spectrum, and the interference factors (f_{yx}, f_{xy}) are obtained experimentally from standard spectra. These two equations may be solved for the two unknowns, either by determinants or substitution, yielding two new equations:

$$X_x = C_1N_x + C_2N_y$$

$$Y_x = C_3N_x + C_4N_y$$

Table 1. Fractional contributions simultaneous equation method

Radionuclide	Channels	Fractional Contribution of Below Listed Radionuclides to the Photopeak of Nuclides Being Assayed*			
		I^{131}	Ba^{140}	Cs^{137}	K^{40}
I^{131}	16-20	1.000	0.866	0.324	0.389
Ba^{140}	23-27	0.025	1.000	0.186	0.256
Cs^{137}	30-36	0.075	0.313	1.000	0.300
K^{40}	70-76	0.0	0.185	0.0	1.000

*Based upon 4" x 4" NaI(Tl) crystal and 3.5 liter water sample in Marinelli beaker.

$$N_i = I_i + f_{ci}C_c + f_{bi}B_b + f_{ki}K_k$$

$$N_c = f_{ic}I_i + C_c + f_{bc}B_b + f_{kc}K_k$$

$$N_b = f_{ib}I_i + f_{cb}C_c + B_b + f_{kb}K_k$$

$$N_k = f_{ik}I_i + f_{ck}C_c + f_{bk}B_b + K_k$$

where

N_i = the net count rate in the photopeak of I^{131}

III. HIGHER ORDER MATRICES

A similar set of simultaneous equations can be written for more radionuclides. Consider I^{131}, Cs^{137}, Ba^{140}, and K^{40}, which are found in liquid milk samples. If these radionuclides are designated as I, C, B, and K respectively and the net count rate (gross count rate minus background, channel-by-channel) is designated as N, the equations obtained would be as follows:

I_i = the count rate contributed to the photopeak of I^{131} by I^{131}.

f_{ci} = the fractional contribution of Cs^{137} to the photopeak of I^{131}.

The subscripts, i, c, b, and k, or combinations thereof, as shown in the above equations are used to denote I^{131}, Cs^{137}, Ba^{140}, and K^{40}, respectively.

For the case being considered (I^{131}, Cs^{137}, Ba^{140}, and K^{40}), the "f" values have been determined experimentally and are presented in Table 1. These values are based on a counting arrangement using a 3.5 liter water sample in a special container and a 4" × 4" NaI(Tl) crystal. Using the data it is possible to solve these equations by the method of determinates.

The following equations are the solutions:

$\underline{I^{131}}$

$$I_i = 1.034\,N_i - 0.178\,N_c - 0.843\,N_b - 0.152\,N_k$$

$\underline{Cs^{137}}$

$$C_c = -0.074\,N_i + 1.071\,N_c - 0.251\,N_b - 0.222\,N_k$$

$\underline{Ba^{140}}$

$$B_b = -0.013\,N_i - 0.205\,N_c + 1.123\,N_b - 0.232\,N_k$$

$\underline{K^{40}}$

$$K_k = -0.002\,N_i - 0.038\,N_c - 0.208\,N_b + 1.043\,N_k$$

IV. COMMENTARY

The method described herein has been used for rapid assay of milk samples with very good results. Application has been applied to other samples with higher number of nuclides present.

The main advantage of using the simultaneous equation method for assay of gamma spectra is its adaptability for use on a computer. This allows for fast results from a sample. In addition, little or no preparation of the sample has to be done.

The main disadvantage to the method is that radionuclides other than those accounted for in the equation cannot be present if the solution is to be valid.

Also, if a significant number of radionuclides are to be considered, the solution of the simultaneous equations becomes exceedingly difficult if not impossible without the use of an electronic computer.

In working with samples of multiple radionuclides present, several errors may be introduced. The first consideration is that of stability and linearity of the instrument.

If two nuclides fall in the same photopeak, various applications may be used to try and separate them. First one should look for other photopeaks associated with the particular nuclide. Next one may study the half-life of the nuclides and separate them this way. If these fail to work a chemical separation may have to be used.

In determining the "f" factors one must try to keep the geometry, density and homogeneity of the sample media constant.

The more "f" factors that have to be introduced, i.e., more nuclides in sample, the higher the error associated with any one value becomes. Therefore it is imperative that good counting statistics be maintained.

Simultaneous Equation Method

(Quantitative Analysis)

I. INTRODUCTION

Many types of environmental samples, which must be analyzed routinely, have a fairly constant radionuclide content which is limited to a specific minimal number of nuclides. Examples are milk contaminated with 10 day or older fallout, 30 day or older atmospheric fallout particulates, or effluents from normal reactor operations if short-lived nuclides are decayed out.

A convenient method for the quantitative analysis of this type of sample is the "simultaneous equation method." This method entails a mathematical approach for eliminating Compton interference from photopeaks. The only requirements for application of this method are:

1) That all nuclides present in the sample are identified.

2) That a different photopeak be present for each nuclide in the sample. This does not exclude the possibility of many overlapping peaks.

II. DEVELOPMENT OF METHOD

A. Interference Factors

The principle of "constant spectral shape" states that the ratio of any portion of a spectrum to another portion is constant and independent of the activity. A useful ratio is that of the photopeak channel(s) of a different nuclide to the photopeak channel(s) of the nuclide whose spectrum is being considered. This ratio, termed an interference factor, is obtained from a standard spectrum of the single nuclide. The significance of this terminology will become apparent shortly.

First consider the spectrum of the nuclide Z (see Figure 1). It is possible to express the count rate of any channel in terms of any other channel. If channel 10 is taken as the reference peak, the count rate in channel 20 can be written as:

$$CR_{20} = f_{(10-20)} CR_{10}$$

where

CR_{20} = count rate in channel 20

CR_{10} = count rate in channel 10

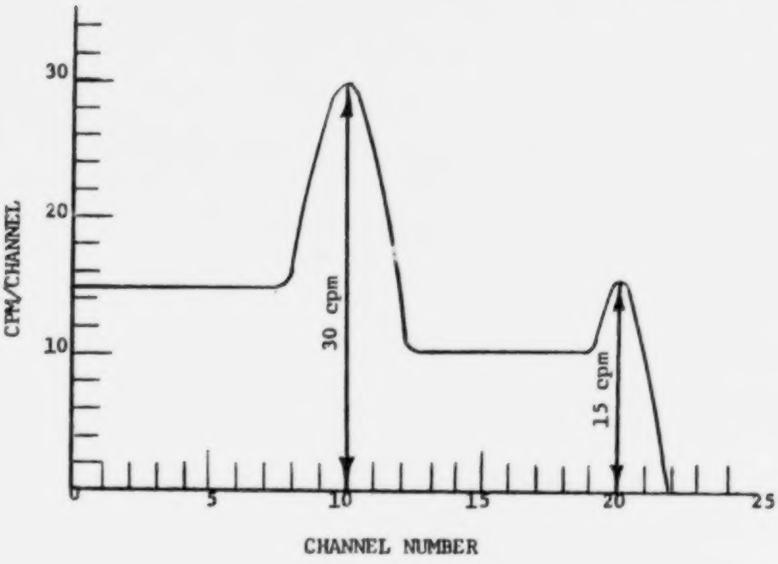
$f_{(10-20)}$ = interference factor

The interference factor is calculated by:

$$f_{10-20} = \frac{CR_{20}}{CR_{10}} = \frac{15}{30} = 0.5$$

thus if count rate in channel 10 of another spectrum of nuclide Z is 50 counts per minute, the count rate in channel 20 becomes 25 cpm.

$$CR_{20} = 0.5 \times 50 = 25 \text{ cpm}$$



SPECTRUM OF NUCLIDE Z

Figure 1

B. Simultaneous Equations

Next the spectrum of a sample containing two radionuclides, X + Y is considered. Figure 2 shows the individual spectra of X and Y and the composite spectrum.

The net count rate of the composite spectrum for the photopeak channel of nuclide X (N_x) can be written as:

$$N_x = X_x + X_y \qquad (1)$$

where

X_x = count rate contributed by nuclide X to the photopeak channel of X

X_y = count rate contributed by nuclide Y to the photopeak channel of X

However, X_y may also be expressed in terms of Y_y, the count rate contributed by nuclide Y to the photopeak channel of Y.

$$X_y = f_{yx} Y_y \qquad (2)$$

where

f_{yx} = interference factor which expresses the fractional contribution of the photopeak channel of nuclide Y to the photopeak channel of nuclide X.

Combining equations (1) and (2), we have:

$$N_x = X_x + f_{yx} Y_y$$

A similar development can be done for the net count rate in the photopeak channel of nuclide Y (N_y), resulting in the equation:

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The two equations:

$$N_x = X_x + f_{yx} Y_y$$

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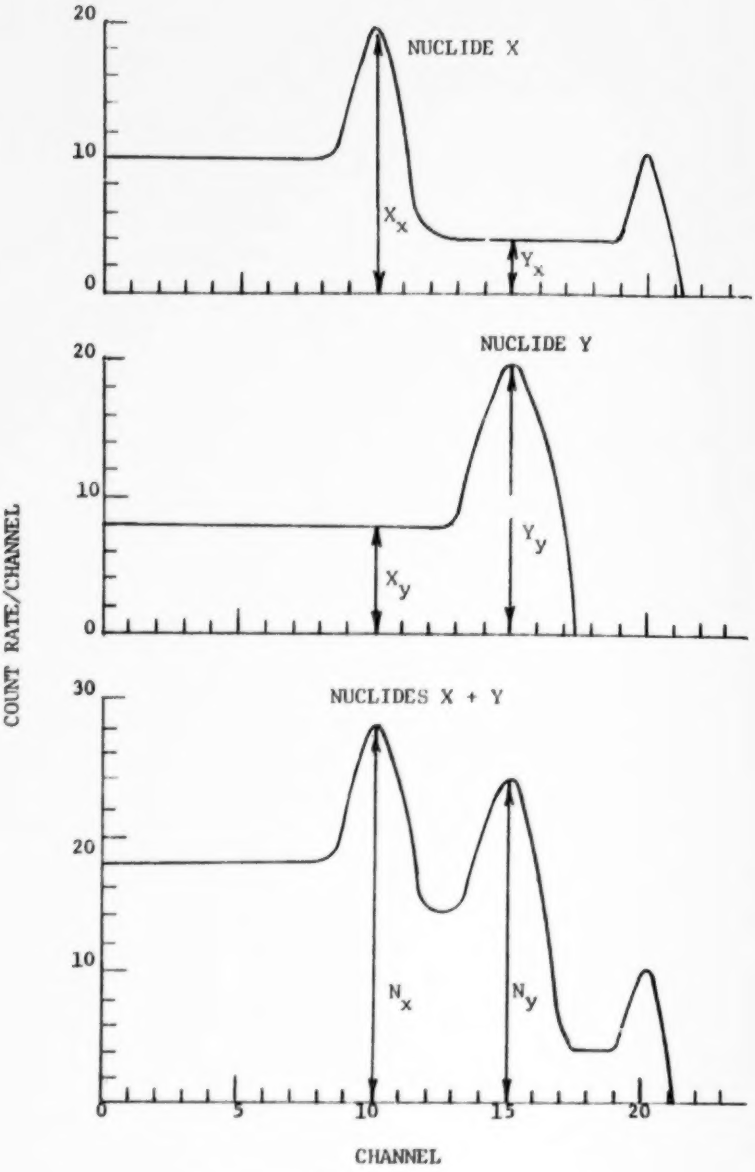

SPECTRUM OF COMPOSITE SAMPLE

Figure 2

are simultaneous linear independent equations containing two unknowns, X_x and Y_y. The net count rates (N_x, N_y) of the composite sample are obtained from the composite spectrum, and the interference factors (f_{yx}, f_{xy}) are obtained experimentally from standard spectra. These two equations may be solved for the two unknowns, either by determinants or substitution, yielding two new equations:

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where

N_i = the net count rate in the photopeak of I^{131}

I_i = the count rate contributed to the photopeak of I^{131} by I^{131}.

f_{ci} = the fractional contribution of Cs^{137} to the photopeak of I^{131}.

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Solution of Simultaneous Linear Equations by Determinants

I. INTRODUCTION

The solution of simultaneous linear equations, such as those developed in determining the activity from gamma spectral data, can be done in a number of ways. One method, to be discussed in this outline, is by use of determinants. A determinant is a square array of numbers, i.e., equal number of rows and columns, and has a numerical value. The number of rows or columns determines the order of the determinant. Therefore, a determinant having two rows and two columns would be a second order determinant; three rows and three columns would be a third order determinant.

II. SOLUTION OF 2nd AND 3rd ORDER DETERMINANTS

The solution of two or three simultaneous linear equations is straight forward and is best explained by use of examples.

EXAMPLE

Given: a system of linear equations

$$a_1x + b_1y + c_1z = d_1$$
$$a_2x + b_2y + c_2z = d_2$$
$$a_3x + b_3y + c_3z = d_3$$

Where:

x, y, z - unknown quantities

a_1, a_2, a_3 - coefficients of unknown x

b_1, b_2, b_3 - coefficients of unknown y

c_1, c_2, c_3 - coefficients of unknown z

d_1, d_2, d_3 - constant terms

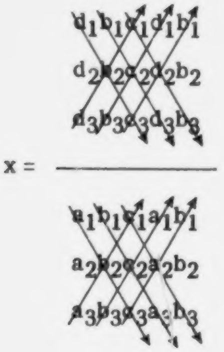
The value of each unknown (x, y, z) is determined by evaluating the following determinants. Note that the determinant of the denominator, in all cases, is the coefficient of each unknown written down in order.

Setting up the determinant of the numerator, for any unknown, is accomplished in the same manner as the denominator, with the additional step of replacing the coefficients of that unknown with the appropriate constant terms.

EXAMPLE

$$x = \frac{\begin{vmatrix} d_1b_1c_1 \\ d_2b_2c_2 \\ d_3b_3c_3 \end{vmatrix}}{\begin{vmatrix} a_1b_1c_1 \\ a_2b_2c_2 \\ a_3b_3c_3 \end{vmatrix}} \quad y = \frac{\begin{vmatrix} a_1d_1c_1 \\ a_2d_2c_2 \\ a_3d_3c_3 \end{vmatrix}}{\begin{vmatrix} a_1b_1c_1 \\ a_2b_2c_2 \\ a_3b_3c_3 \end{vmatrix}} \quad z = \frac{\begin{vmatrix} a_1b_1d_1 \\ a_2b_2d_2 \\ a_3b_3d_3 \end{vmatrix}}{\begin{vmatrix} a_1b_1c_1 \\ a_2b_2c_2 \\ a_3b_3c_3 \end{vmatrix}}$$

Each determinant is expanded by repeating the first two columns and multiplying each diagonal set of three coefficients. The product is given a positive (+) sign for sets running down to the right and a negative (-) sign running up to the right.



$$x = \frac{\begin{matrix} d_1 b_1 c_1 d_1 b_1 \\ d_2 b_2 c_2 d_2 b_2 \\ d_3 b_3 c_3 d_3 b_3 \end{matrix}}{\begin{matrix} a_1 b_1 c_1 a_1 b_1 \\ a_2 b_2 c_2 a_2 b_2 \\ a_3 b_3 c_3 a_3 b_3 \end{matrix}}$$

which upon expansion takes the form

$$x = \frac{d_1b_2c_3 + b_1c_2d_3 + c_1d_2b_3}{a_1b_2c_3 + b_1c_2a_3 + c_1a_2b_3}$$

$$\frac{- d_3b_2c_1 - b_3c_2d_1 - c_3d_2b_1}{- a_3b_2c_1 - b_3c_2a_1 - c_3a_2b_1}$$

The solution for y and z can be determined the same way. It is of interest to note that the denominator will be the same for all the unknowns, x, y, z.

The solution for a set of two equations can be determined in the same manner as above. In this case, two equations, you would have only x, y unknowns, a_1, a_2, b_1 and b_2 coefficients, and d_1 and d_2 constant terms.

III. DETERMINANTS OF FOURTH ORDER

As seen, the solution of two or three simultaneous equations is straight forward. However, when we have more than three equations, the solution becomes more involved.

A few terms should be understood before we discuss the solution of nth order determinants.

A. Minor - in any determinant, if the row and column containing a given element, say E, are blotted out, the determinant formed from the remaining elements is called the minor of E.

EXAMPLE

In $\begin{vmatrix} a_1b_1c_1 \\ a_2b_2c_2 \\ a_3b_3c_3 \end{vmatrix}$ the minor of c_2 is $\begin{vmatrix} a_1b_1 \\ a_3b_3 \end{vmatrix}$

B. Cofactor - a cofactor is a minor with a sign (±) attached. The signs to be attached to the minors of the elements of a determinant in order to obtain the cofactors, can be determined by use of Figure 1.

Figure 1

$$\begin{vmatrix} + & - & + & - & . & . & . \\ - & + & - & + & . & . & . \\ + & - & + & - & . & . & . \\ - & + & - & + & . & . & . \\ . & . & . & . & . & . & . \end{vmatrix}$$

The sign in each location is the one which should be attached to the minor of the corresponding element in expanding. The signs alternate in preceding to the right in any row, or moving down in any column.

EXAMPLE

In $\begin{vmatrix} a_1 b_1 c_1 d_1 \\ a_2 b_2 c_2 d_2 \\ a_3 b_3 c_3 d_3 \\ a_4 b_4 c_4 d_4 \end{vmatrix}$ the cofactor of a_3 is

$+ a_3 \begin{vmatrix} b_1 c_1 d_1 \\ b_2 c_2 d_2 \\ b_4 c_4 d_4 \end{vmatrix}$ the cofactor of a_4 is $- a_4 \begin{vmatrix} b_1 c_1 d_1 \\ b_2 c_2 d_2 \\ b_3 c_3 d_3 \end{vmatrix}$

With those terms in mind, let us examine how a set of four simultaneous equations can be solved.

EXAMPLE

Given: a set of linear equations

$$a_1 v + b_1 x + c_1 y + d_1 z = f_1$$
$$a_2 v + b_2 x + c_2 y + d_2 z = f_2$$
$$a_3 v + b_3 x + c_3 y + d_3 z = f_3$$
$$a_4 v + b_4 x + c_4 y + d_4 z = f_4$$

Where:

$v, x, y, z,$ - unknown elements

a_1, a_2, a_3, a_4 - coefficients of unknown v

b_1, b_2, b_3, b_4 - coefficients of unknown x

c_1, c_2, c_3, c_4 - coefficients of unknown y

d_1, d_2, d_3, d_4 - coefficients of unknown z

f_1, f_2, f_3, f_4 - constant terms

The value of each unknown (v, x, y, z) is determined by evaluating the following determinants.

$$v = \frac{\begin{vmatrix} f_1 b_1 c_1 d_1 \\ f_2 b_2 c_2 d_2 \\ f_3 b_3 c_3 d_3 \\ f_4 b_4 c_4 d_4 \end{vmatrix}}{\Delta}$$

$$x = \frac{\begin{vmatrix} a_1 f_1 c_1 d_1 \\ a_2 f_2 c_2 d_2 \\ a_3 f_3 c_3 d_3 \\ a_4 f_4 c_4 d_4 \end{vmatrix}}{\Delta}$$

$$y = \frac{\begin{vmatrix} a_1 b_1 f_1 d_1 \\ a_2 b_2 f_2 d_2 \\ a_3 b_3 f_3 d_3 \\ a_4 b_4 f_4 d_4 \end{vmatrix}}{\Delta}$$

$$z = \frac{\begin{vmatrix} a_1 b_1 c_1 f_1 \\ a_2 b_2 c_2 f_2 \\ a_3 b_3 c_3 f_3 \\ a_4 b_4 c_4 f_4 \end{vmatrix}}{\Delta}$$

where $\Delta = \begin{vmatrix} a_1 b_1 c_1 d_1 \\ a_2 b_2 c_2 d_2 \\ a_3 b_3 c_3 d_3 \\ a_4 b_4 c_4 d_4 \end{vmatrix}$

The solution of the numerator for v is as follows:

$$\begin{vmatrix} f_1b_1c_1d_1 \\ f_2b_2c_2d_2 \\ f_3b_3c_3d_3 \\ f_4b_4c_4d_4 \end{vmatrix} = +f_1 \begin{vmatrix} b_2c_2d_2 \\ b_3c_3d_3 \\ b_4c_4d_4 \end{vmatrix} - f_2 \begin{vmatrix} b_1c_1d_1 \\ b_3c_3d_3 \\ b_4c_4d_4 \end{vmatrix}$$

$$+f_3 \begin{vmatrix} b_1c_1d_1 \\ b_2c_2d_2 \\ b_4c_4d_4 \end{vmatrix} - f_4 \begin{vmatrix} b_1c_1d_1 \\ b_2c_2d_2 \\ b_3c_3d_3 \end{vmatrix}$$

The denominator Δ can be solved in same manner.

$$= \Delta \begin{vmatrix} a_1b_1c_1d_1 \\ a_2b_2c_2d_2 \\ a_3b_3c_3d_3 \\ a_4b_4c_4d_4 \end{vmatrix} + a_1 \begin{vmatrix} b_2c_2d_2 \\ b_3c_3d_3 \\ b_4c_4d_4 \end{vmatrix} - a_2 \begin{vmatrix} b_1c_1d_1 \\ b_3c_3d_3 \\ b_4c_4d_4 \end{vmatrix}$$

$$+ a_3 \begin{vmatrix} b_1c_1d_1 \\ b_2c_2d_2 \\ b_4c_4d_4 \end{vmatrix} - a_4 \begin{vmatrix} b_1c_1d_1 \\ b_2c_2d_2 \\ b_3c_3d_3 \end{vmatrix}$$

The third order determinants can be solved by the previously described method or by further resolving the 3 by 3 determinant to a 2 by 2 determinant with the use of cofactors. The solution of x, y, z would be similar to the method described for v.

IV. HIGHER ORDER DETERMINANTS

The problem of solving higher order determinants becomes more time consuming. Where a 4th order determinant may take from 1 to 2 hours, a 5th order determinant may take from 4 to 6 hours. Therefore, for 6th order and higher determinants, it is most practical to resort to computer techniques.

REFERENCE

Kaplen, Wilfred. Advanced Calculus. Addison-Wesley Publishing Company, Inc. Reading, Massachusetts. 1959.

Instrumental Data Manipulations

I. INTRODUCTION

A complete analysis of a gamma spectrum requires the analyst to go through many types of techniques. Such techniques might include visual observance of the plotted spectrum; multiplication, division, addition, and subtraction of sample spectrum and standard spectrum data; and the use of certain instrumental capabilities to manipulate spectral data of interest. These operations may be required where the initial information from the gamma spectrometer is not in the final qualitative and quantitative form desired by the analyst.

This outline will discuss certain instrumental data manipulations or data collections that are of value in the analysis of gamma spectra of environmental samples. This approach is greatly facilitated by the wide flexibility afforded the analyst in modern-day gamma analyzers.

II. TIMER MECHANISMS

Most spectrometer systems have a timer mechanism either as an auxiliary unit or as an integral part of the system itself.

A. Value of a Timer

Environmental samples have to be counted for sufficient periods of time in order to obtain adequate counting statistics. The length of the counting time may vary for different samples due to considerations such as sample radioactivity content, instrument background, desired precision, etc. Therefore, it is advantageous to have the capability of presetting the timer for variable time intervals.

Many advantages are afforded by a flexible timer mechanism. If the timer can be set for a definite counting time and the data collection will automatically stop at the end of this time, it does not require the analyst to be present at the end of the counting period. Some timers have a recycling capability in which the following sequence occurs: 1) the sample is counted for a preset time interval as determined by the analyst, 2) the information is "read out", 3) the sample is automatically recounted for the same time interval, and 4) the information is "read out". This sequence can be repeated as long as desired. This capability allows the analyst to gather information concerning a sample counted several times, and it does not require him to be present to operate the spectrometer. One application of the information gathered would be the half-life determination of short-lived radionuclides.

Another useful timer flexibility is the ability to stop the timer while counting, view the spectrum obtained to that point, and then continue the sample count. By viewing the spectrum any unusual occurrences could be noted and corrected where possible, (i.e., gain shift could be noted, corrected, and the count completely restarted) or a rough check as to whether the memory capacity is sufficient could be made.

B. Dead and Live Time

The spectrometer requires a certain period of time (in the order of microseconds) to analyze a pulse from the detector and store this information. During the period of time in which the pulse is being analyzed and the resulting information stored the spectrometer system cannot accept another pulse, thus, this time is known as instrument "dead" time.

The time during which the instrument can actually accept a pulse is known as instrument "live" time. Therefore, the actual "clock" time minus the dead time equals the live time.

In comparing a sample spectrum with a standard spectrum for a radionuclide it is important that the live times be related. In some environmental analyses the dead time of the instrument is insignificant and this consideration is of no consequence. For higher activity environmental samples however, this consideration may be important.

Different spectrometer systems may have different means of indicating live time, included among these are the following: 1) direct live time indication on a timer register (this is accomplished by having a register for clock time and live time, or by having one register with a switch to allow the selection of either clock or live time), or 2) indirect live time indication as a percent of actual clock time (this is accomplished by having a register indicating percent live time and another register indicating actual clock time).

In some instruments the dead time is variable with respect to the channel in which the count is to be stored; therefore, a fixed dead time switch may be provided which will compensate for the variations. A fixed dead time switch allows the analyst to set a constant dead time which is greater than the longest dead time of the analyzer. The setting of a constant dead time is of value when the instrument has no live time indicator.

III. TEMPORARY ANALOG STORAGE

Some instruments have the capability of receiving the second of a pair of closely spaced pulses even though the analyzer has not completed the analysis of the first pulse when the second pulse occurs. The pulses are stored in a pulse stretcher when the analyzer is busy and upon completion of the preceding pulse analysis, the second pulse proceeds.(1) This type of operation essentially increases the "live" time (decreases "dead" time) of the instrument, thus, improving counting efficiency.

IV. MEMORY FLEXIBILITY

One of the most important and most flexible portions of a gamma spectrometer system is the memory. Many data manipulations are performed using the memory and/or accessory units which operate on the data contained in the memory. In some multi-channel systems the memory can be divided into two, four, or more sections, and data can be transferred between sections. Data transfer (data transfer is accomplished when all of the information in one subgroup is transferred on a channel-by-channel basis to another subgroup) offers advantages in analysis, and memory splitting allows the use of multiple input detectors and/or input subgroupings.

A. Data Transfer

Data transfer is a very important step in visual comparison of spectra, and in background subtraction using subgroups from one input.

Some instruments have the capability of bi-directional transfer (transfer to either higher or lower numbered channels), while others only permit data transfer in one direction (transfer from higher to lower numbered channels and not vice versa).

In most instruments, data transfer is initiated manually by the analyst; however, some instruments also provide automatic transfer. Automatic data transfer is of value when subtracting background using the recycling timer mechanism.

B. Multiple Inputs

When more than one detector is used in conjunction with a pulse analyzer and readout system many advantages are afforded. One obvious advantage is that more samples could be counted over a period of time; however, massive shielding would be required for each detector if environmental samples were counted.

The capacity of the memory and the number of sections into which it can be divided is a limitation to the capacity of each input. For example - if the memory had a capacity of 400 addresses (channels) and it could be divided into four equal sections, four inputs with a capacity of 100 channels each could be used; if the same 400 channel memory could only be divided into two equal sections, two inputs could be used and each would have a capacity of 200 channels.

C. Input Subgroups

The ability to subdivide the memory core into various sections does not necessarily mean that multiple inputs should be provided. Actually, the ability to divide the memory into sections (subgroups) from one input can also be advantageous. For example: the analyst may count a sample on one input in one, two or four hundred channels if the memory can be divided into four sections. By choosing the number of channels to be used in counting a sample the analyst can control the energy range of the spectrum, with the channel coefficient being dependent on the instrument gain setting. There are many other uses of input subgrouping, some of which will be discussed subsequently.

V. APPLICATIONS OF MEMORY FLEXIBILITY

The uses of memory flexibility in analyzing gamma spectra depend mainly on two features of the memory: 1) the ability to divide the memory into sections, and 2) the ability to transfer data from section to section.

A. Background Subtraction

One advantage offered by input subgrouping is instrument background subtraction. This operation can be accomplished when the instrument has the capability of either direct counting in the negative mode or complementing a number with respect to the memory capacity. Also depending on the memory section being used, data transfer may or may not be required. These capabilities will be discussed in more detail subsequently.

The advantages offered by instrumental background subtraction are mainly related to spectral analysis. Upon simplification of a spectrum by background subtraction the qualitative identification and quantitative evaluation can be more easily accomplished.

Actually, the real distinguishing feature of various instruments in relation to background subtraction is the method of getting the background data in the negative form in the channel subgroup of interest. These methods will be discussed in terms of direct negative mode counting, complementation, and magnetic tape background subtraction.

1. DIRECT NEGATIVE MODE COUNTING

In negative mode counting the counts detected by the instrument are subtracted from the memory capacity on a channel-by-channel basis and the resulting number is recorded in the memory. Getting information into the

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desired memory location may be accomplished by counting in the negative mode in the subgroup of interest, or by counting in the negative mode in another subgroup and transferring the data.

A sequence of operation for background subtraction would be as follows: 1) collection of background data in the negative mode in the channel subgroup of interest, and 2) counting of sample in positive mode in the channel subgroup of interest, with the resulting information in the memory being due only to those counts caused by the sample itself.

The following serves as a numerical example of direct negative mode counting: if the memory capacity is 99,999 counts per address (channel) and 292 counts due to background are detected in channel 1, the number actually entered in the memory for channel 1 would be 99,707 (99,999-292). (When the sample is counted we assume that the background still contributes 292 counts and that all counts above this are due to the sample.) If sample plus background counting yields 500 counts for that channel, the actual sample count is 208 (500-292). The 500 counts are collected in the positive mode and added to the 99,707 counts already stored there from the background count. When the memory capacity is exceeded in any one channel that channel's data reverts to 0 counts and begins again. Therefore, when 293 of the 500 sample plus background counts have been registered, the channel will read 1 count 99,707 + 293 = 100,000 (1 count over capacity)

One evident disadvantage of direct negative mode counting in the channel subgroup of interest is that a background count must be taken for each sample. When the direct negative mode counting is done in another subgroup and the data is non-destructively transferred, a single background count could suffice for several sample countings.

1. COMPLEMENTATION

The complementation method of subtracting background has been reported(2) with the following sequence of operations. The sample plus background is counted in the positive mode in the subgroup of interest, and the complement of the resulting count is taken with respect to the channel memory capacity. This occurs by pushing a button which causes the number to be complemented to be read from the memory into an increment adder and subtracted from the channel capacity. Then this resulting number is returned to the memory, this number being the same as would be obtained by direct counting in the negative mode. The background is then counted in the positive mode in the same subgroup, then the complement of the resulting number is taken with respect to the channel capacity. The resulting number will be equal to the counts due to the sample alone. The instrument would perform this operation on a channel-by-channel basis.

An example of this operation is as follows:

Assume channel capacity = 99,999 counts.

Assume that upon counting sample plus background a count of 12,248 is obtained in channel 10. Upon taking the complement of this number with respect to the channel's capacity, 87,751 (99,999-12,248) is obtained.

Upon counting background a count of 6421 is obtained, this number is instrumentally added to the 87,751 counts already present in channel 10, yielding 94,172. Upon taking the complement of this count (94,172) with respect to the channel capacity (99,999), 5827 counts is obtained, this number being the same as would be obtained by subtracting the background from the sample plus background (12,248-6421 = 5827).

3. MAGNETIC TAPE BACKGROUND SUBTRACTION

Background counts may also be gotten into the desired memory location in the negative form by "reading" information into the subgroup of interest from a magnetic tape "storage". Information from tape storage could also be "read" into another subgroup and the data transferred to the subgroup of interest.

The information on the magnetic tape may be the result of a composite background averaged over a long period of time. In order to get the proper information on a magnetic tape the following steps would have to be followed:

1) Collect several backgrounds in the negative mode and print the information.

2) Enter this information into a computer and allow the computer to average the negative mode background count for each channel, calculate the counts for each channel for the time period the sample is to be counted, and store this information on a magnetic tape.

3) Enter the background information into the desired memory subgroup by transferring the information on the magnetic tape through a tape reader to the memory location.

After the background data is gotten to the subgroup of interest, either by directly entering or by data transfer, the sample plus background is counted in the positive mode, thus allowing the instrument to subtract background in the manner previously discussed.

This method would be of value for instruments having a stable background because it would not require frequent background countings.

B. Visual Spectral Comparison

Quite often in gamma spectral analysis it is necessary to visually compare spectra. By knowing the relative spectral shapes of samples and standards the subsequent qualitative and quantitative analysis may be facilitated.

If the spectrometer has multiple inputs and "live" display capabilities a visual comparison between the spectrum from each input can be made. This operation would be of value in comparing a sample with a background count, a standard and a sample, or two samples. There are many other possibilities for the use of visual comparison of data from multiple inputs, some of which would be dependent upon the function of the particular installation.

Visual spectral comparison of data from subgroups of one input can also be of value. The same comparisons as previously mentioned could be made.

A disadvantage of visual spectral comparison without the use of external magnetic tape storage is that spectra being compared would have to be counted at the time of comparison. However, with magnetic tape storage, data from a background spectrum and "standard" spectra could be "stored" and then "read" into the memory prior to visual comparison. The data would be read into the memory in one section and then visually compared by live display to some other data in another section. This operation would greatly facilitate the total time required for spectral comparison, and it could be used for both multiple input capabilities, multiple input - input subgrouping capabilities, and single input - input subgrouping capabilities.

C. Integration

In qualitative identification of gamma spectra using spectrum stripping, and in quantitative evaluation using the Compton Continuum method, spectrum stripping, or the simultaneous equation method, the counts in various channels have to be summed. Several present-day multi-channel analyzers have the capability of instrumentally integrating the counts in selected channels and yielding the summed result.

There are two modes of integration normally found in spectrometer systems-the "normal" mode and the "sub-total" mode. The normal mode of integration occurs when the instrument internally integrates the counts in the selected channels and the resultant total is displayed. The sub-total mode occurs when the total counts of the integrated channels is indicated for each successive channel addition.

One very important aspect of integration is the selection of the proper channels. Visual observance of the spectrum may be an aid in this selection.

D. Spectrum Stripping

Spectrum stripping (removal of the contribution of a higher energy gamma photon from a photopeak resulting from a lesser energy gamma photon) can be a very important operation in both the qualitative and quantitative analysis of a spectrum. The basic principles applied in spectrum stripping are as follows: 1) under given physical counting conditions, the counting of a radionuclide will yield a spectrum of constant relative spectral shape, irrespective of activity, and 2) the area under a spectrum for a single nuclide, or any portion of the area, is proportional to the activity of the radionuclide.

Spectrum stripping may be performed manually on the entire spectrum or on any portion thereof.

Commercial spectrum stripping instruments are available which, when used in conjunction with the gamma spectrometer memory core, perform spectrum stripping semi-automatically.

The commercial spectrum stripper is essentially composed of an external storage memory (magnetic tape, punched paper tape, etc.) which contains normalized standard spectra; and logic circuitry which is capable of multiplication and subtraction. The basic sequence of instrumental spectrum stripping is as follows:

1. The person performing the analysis manually determines a spectrum multiplier and enters it by key punch into the instrument along with a code designation for the standard spectrum to be subtracted.

2. The external storage memory is scanned to find the particular spectrum needed.

3. Channel 1 is read-out of the spectrum storage memory and operated on by the spectrum multiplier. At the same time, channel 1 of the analyzer memory is read-out and the adjusted standard is subtracted from it. These operations are performed in the spectrum stripper logic circuitry.

4. The subtracted channel 1 count information is then re-entered into the memory of the analyzer.

5. The same process is repeated for each channel until all channels are subtracted.

Actually, most present-day spectrum strippers do not have the capability of multiplying the normalized standard spectra by the spectrum multiplier on a one-operation basis. Multiplier capability usually ranges over several multiples of 10; e.g., ×10, ×1, ×0.1, ×0.01, and ×0.001. An example illustrates the application of these various multipliers:

Assume the spectrum multiplier is 1.58 for the standard of interest. To multiply each channel count rate of the standard by 1.58 and then subtract these values channel-by-channel from the sample spectrum being analyzed the following steps would have to be performed:

1) Punch × 1 multiplier into spectrum stripper and the code for standard spectrum of interest (instrument carries out subtraction operation).

2) Punch ×0.1 multiplier into spectrum stripper and the code for the standard

spectrum of interest (instrument carries out subtraction process)

3) Repeat step 2) four more times.

4) Punch × 0.01 multiplier into spectrum stripper and the code for the standard spectrum of interest (instrument carries out subtraction process)

5) Repeat step 4) seven more times.

It should be noted that the multiplication does not have to be performed in prescribed sequence.

Even though the semi-automatic instrumental stripping may require several operations, it is still faster, timewise, than performing spectrum stripping manually.

E. Synthesization of Spectra

In some situations it may be desirable to synthesize a spectrum from several standard spectra and then compare this with the sample spectrum being analyzed.

The spectrum to be synthesized would be "built up" in a cleared input subgroup. The basic steps of synthesization would be the same as in spectrum stripping, except that the data would be added as it enters rather than subtracted.

F. Successive Counting

Some analyzers have the capability of counting in one subgroup of an input for a period of time and then automatically moving to the next subgroup and counting for the same period of time without any intermediate data readout.(2) Other analyzers can accomplish the same operation if the analyst manually switches the subgroup and resets the timer after the first count. The advantage offered by counting in successive subgroups is that short-lived radionuclides can be analyzed more closely without having to take the time for readout.

Some analyzers also have the capability of counting gross (without respect to energy) counts in one channel for a short time period and then continuing this operation on a channel-by-channel basis. The time allotted to each channel may be referred to as the "dwell" time. Actually, the operation essentially converts the abscissa from channel number to time, thus allowing a readout of gross counts versus time. This capability would be very valuable in the analysis of short-lived radionuclides.

G. Destruction of Memory Data

At the end of a particular operation or sample counting, and after the desired readout has been obtained, the data in the memory is destroyed (memory cleared). On most instruments, memory clearing may be accomplished manually or automatically following some other instrument function.

VI. SUMMARY

The qualitative and quantitative evaluation of gamma spectra may involve many operations. Some of the analysis operations can be greatly facilitated by instrument flexibilities. Timer features, temporary analog storage features, and memory features serve as examples of certain instrument capabilities the analyst may apply. Operations involving the use of the memory include background subtraction, visual spectral comparisons, integration, spectrum stripping, spectrum synthesization, and successive counting.

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1. IRE Transactions Nuclear Science, NS-9, Eighth Scintillation Counter Symposium, p. 276.

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Application of the Single-Channel Analyzer to Gamma Spectroscopy

I. INTRODUCTION

The use of the multichannel analyzer has facilitated the development of precise and complex qualitative and quantitative analysis techniques. With some limitations these same analyses can be done with a single-channel analyzer, and in certain cases its use may be preferred. The most significant overall advantage of the single-channel analyzer is its lesser cost in comparison with the multichannel analyzer.

This outline will discuss the capabilities of the single-channel analyzer and its application to both qualitative and quantitative analysis.

II. COUNTING TECHNIQUE

The simplest type of counting uses a voltage discriminator to cut out low voltage pulses and a scaler to record all pulses exceeding this minimum voltage. Gross counting (alpha, beta, or gamma) is accomplished in this manner.

To determine the relative contributions of different energy emissions, the voltage discrimination must be adjustable (1, 2, 3, & 4) and the pulse size must be proportional to the initial radiation energy.

A. The Integral Spectrum

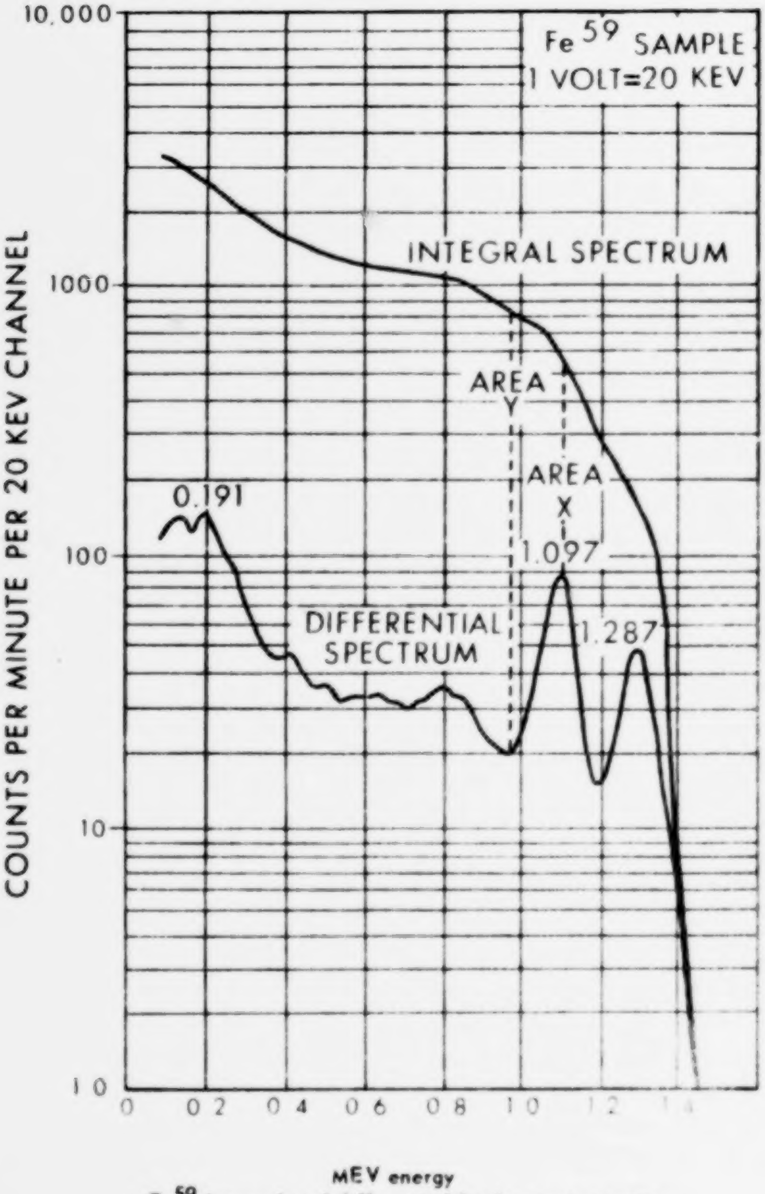
The simplest types of gamma energy analysis is accomplished by using an adjustable baseline discriminator and a scaler. The discriminator is set and a count is made of all pulses having energies with voltage equivalents greater than the baseline. All pulses less energetic are rejected. The baseline discriminator is then moved upward and another count is made of all pulses having energies greater than this baseline setting. The process is repeated until all pulses are being rejected. The net integral spectrum that one would see for a Fe^{59} sample is given in Figure 1.

The distribution of counts at each energy increment can be obtained by differentiating the integral point-by-point. This curve is given in Figure 1. The points on the integral spectrum that have a slope approaching zero coincide with points in the differentiated spectrum where relatively few pulses are occurring. (See Figure 1A and 1B, area marked Y). The slope of the integral spectrum is greatest at the peaks of the differentiated spectrum, where the greatest number of pulses occur (See figure 1, area marked X). Typical integral and differential background spectra are shown in Figure 2.

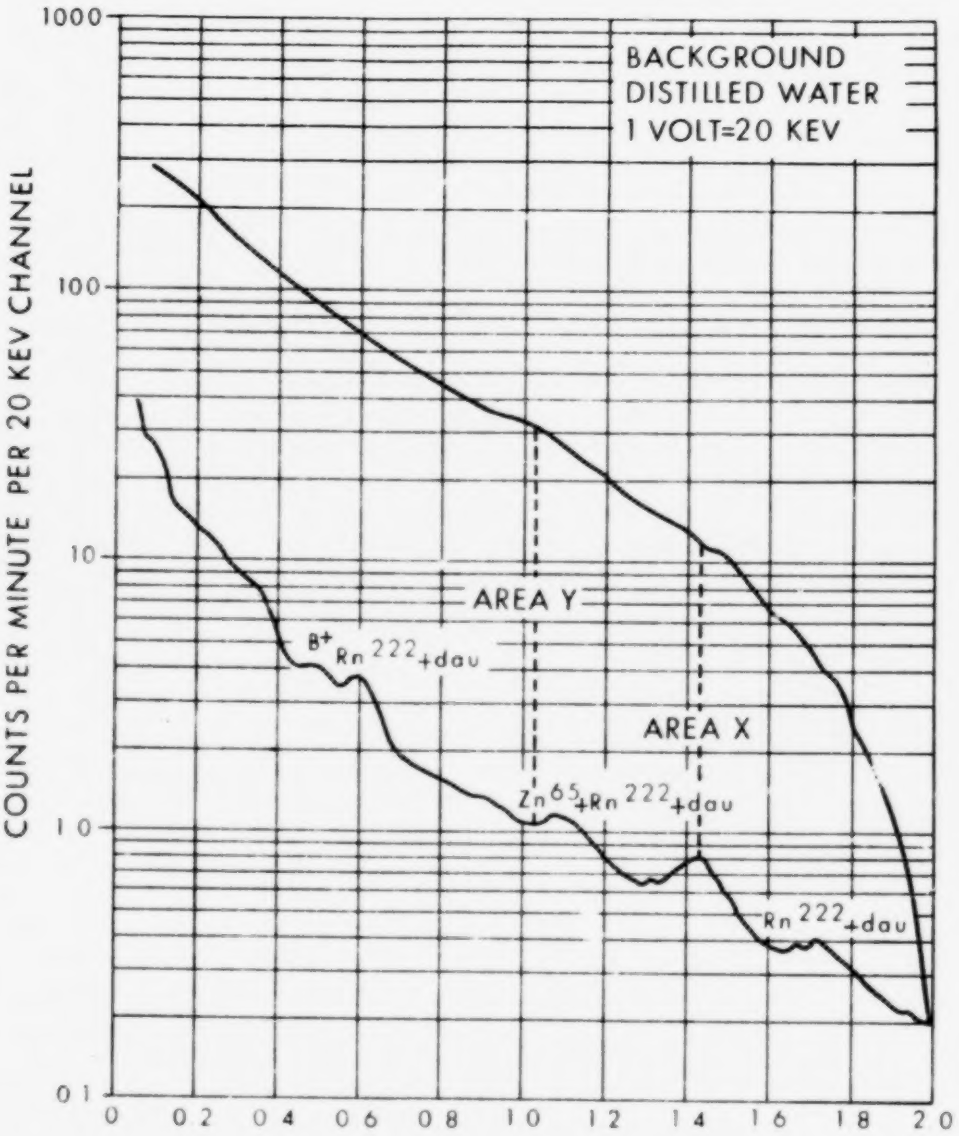
B. The Differential Spectrum

Instrumental techniques have been developed for differentiating the integral spectrum using both a lower and an upper level discriminator. The difference between the discriminators, which is adjustable, is called the window width.

One method of determining the number of pulses that fall between the two discriminators is to count the number of pulses exceeding each discriminator level with a separate scaler and use the difference. This technique involves subtracting one large



Fe^{59} Integral and differential background spectra
Figure 1



MEV energy
Integral and differential background spectra
Figure 2

number to get a small difference. The relative statistical error from such an operation is large.

A second technique which is most commonly used, uses an anti-coincidence circuit to cancel all pulses that exceed the upper discriminator level or fall below the lower discriminator level. Only one scaler of lower capacity is required and manual subtraction is avoided.

C. Comparison of Differential and Integral Counting

Differential counting is commonly used for the energy calibration of a single-channel analyzer and for qualitative analysis.

The integral mode of counting may have advantages in certain applications such as the determination of drift. The region of maximum slope in the integral spectrum corresponds to the photopeak of the differential spectrum (See Figure 1A and 1B, area X). The lower level discriminator is set at the midpoint of the maximum slope portion so that any drift in the baseline will cause a large change in count rate. The formula for determining drift is given in equation (1).

$$\text{Drift in volts} = \frac{\text{change in count rate (cpm)}}{\text{slope of integral spectrum (cpm/volt)}} \text{ or}$$

$$D = \frac{N_0 - N}{\text{slope}} = \text{volts} \qquad (1)$$

where

N_0 = count rate at time t_1

N = count rate at time t_2

The slope is obtained by approximating the region of greatest slope of the integral spectrum with a straight line and calculating the change in count rate with voltage. The drift can be expressed in terms of volt per unit time such as volts/min. or volts/day. If the calculated value of drift is positive the baseline is effectively shifted upwards; if negative, downwards. The advantage of an integral drift determination lies in the better counting statistics. To use differential counting, a window is set on one of the sides of the peak. For sensitivity the window must be narrow and the small number of counts in such a narrow window would have poor counting statistics. The window width stability can be evaluated using differential counting after using the integral counting technique for baseline drift.

The choice between differential or integral counting must be made for each particular situation. Some of the factors influencing this choice will become evident in the following discussions.

III. QUALITATIVE ANALYSIS

Qualitative gamma analysis (1, 5, 6) is accomplished in the same manner as with a multichannel analyzer. The acquisition of information for each area of the spectrum is done separately rather than simultaneously so that the time required to accumulate the same number of counts as a multichannel analyzer in each area or window setting is simply the number of energy divisions times as great. For a 100 energy division analysis the time required would be 100 times greater than for a 100 channel analyzer.

For sufficient activity levels or when long counting times are available, the single-channel analyzer can do a reasonable job with much less expensive equipment.

A. Differential Analysis Equipment

The simplest analysis technique is to count a small increment of the energy range (say 1 volt out of 100 volts), record the counts, move the window up one volt, count again, and repeat until measurements are made for each increment. Each voltage increment is equivalent to an energy span in Mev. This equivalency is variable and depends on the instrument gain. Thus, the 100 volt span may be equivalent to 0-1 Mev or 0-2 Mev or 0-4 Mev, etc. This procedure is time consuming (> 100 minutes for 100 one minute counts) and tedious. In addition, it is hard to prevent overlaps or gaps between successive windows, that is, counting a small area twice or skipping a small area completely.

Several types of scanners have been developed to scan the spectrum mechanically. Instead of using separate upper and lower level discriminators to obtain the window, the upper level discriminator is replaced by an adjustable window width discriminator tied directly to the lower level baseline discriminator. A 60 cycle electric motor is used to continuously change the baseline discriminator at a constant speed. The window then automatically moves across the spectrum.

Such a system replaces the baseline voltage units with time units. However, the time units are directly convertible to voltage and then to energy as Kev or Mev.

The automatic scanner frees the analyst from the necessity of changing operating conditions manually. The continuous scanner also reduces the problem of precise window placement which is present in manual or automatic step scanners.

The automatic step scanner looks at a spectrum increment for a selected period of time, the window is then moved so that it looks at the next higher energy increment for the selected time period. Precise window placement is required to avoid any gaps or overlaps in the spectrum. Although this stability requirement is perhaps a disadvantage over a continuously moving window the automatic step scanner can be used advantageously in some quantitative applications.

In conjunction with the continuous scanner, a ratemeter is used to monitor the number of pulses passed by the single channel analyzer. The ratemeter averages the count rate for a period of time dependent on the time constant and reads out this average cpm. The ratemeter may have either a linear or logarithmic response output. A linear-log output is preferable as both definite applications.

The ratemeter is generally equipped with an adjustable time constant (it can average the count rate over different periods of time), a rate indicating needle to give visual indication of the count rate, and perhaps an aural system. The visual and aural indicators are particularly useful for manual energy calibration of the analyzer.

The ratemeter output is usually fed into a strip-chart recorder. The strip-chart recorder uses time as the abscissa and count rate as the ordinate. The time base of the recorder should be correlated to that of the scanner and ratemeter to obtain conventional spectra. A useful device for this purpose is a mechanism that will cause full-scale deflection of the recorder pen at present baseline voltages (say every 5 volts or half revolution of a 10 turn potentiometer).

B. Differential Analysis Techniques

Matching of scanning speed, ratemeter time constant, and strip-chart recorder paper speed allows direct evaluation of the output in terms of energy for a calibrated analyzer. For low activity samples, the scanning speed should be quite slow giving a long total scan time, the ratemeter time constant long to match it, and the recorder paper feed rate slow to match the scanning speed.

For higher activity samples, the scanning speed may be greater and the ratemeter time constant would be shorter to match the scanner speed or rapid changes in activity levels (peaks versus valleys) will be smoothed out and distorted.

If a calibration signal, say at each 5 volts of the scan is actuated, an non-matching of outputs or slippage in the scanner drive mechanism can be readily detected.

Variable scanner speeds, ratemeter time constants, and strip-chart recorder speeds enables better matching of the scans conditions to the activity of the sample.

The window width plays an important part in the identification of nuclides from spectra. Figure 3 shows the spectrum deformation induced by using three (3) and five (5) volt windows versus a one (1) volt window.

The five volt window is still small compared to the peak width but resolution (separation of peaks) is deteriorating due to the larger energy span covered by the window.

The use of a time constant on the ratemeter longer than the effective scanning time constant will display the same effect as a wide window. The optimum window width and scanning time combination for qualitative analysis would be one with which the counting statistics are improved as much as deteriorating resolution will permit.

C. Integral Spectral Analysis

An integral spectrum can be used for both qualitative and quantitative analysis. One such application (7) has been developed by HASL (Health and Safety Laboratory, U.S.A.E.C., New York). In this case a graphical technique is used to resolve the activity of up to four component mixtures. The process involves integral spectrum stripping and has been developed for low activity samples such as environmental milk samples and whole body counting. By using total counting rates rather than photopeak counting rates improved counting statistics are obtained. In contrast to differential spectrum stripping techniques, there can be no negative numbers generated.

IV. QUANTITATIVE ANALYSIS

A. Single Nuclide Counting

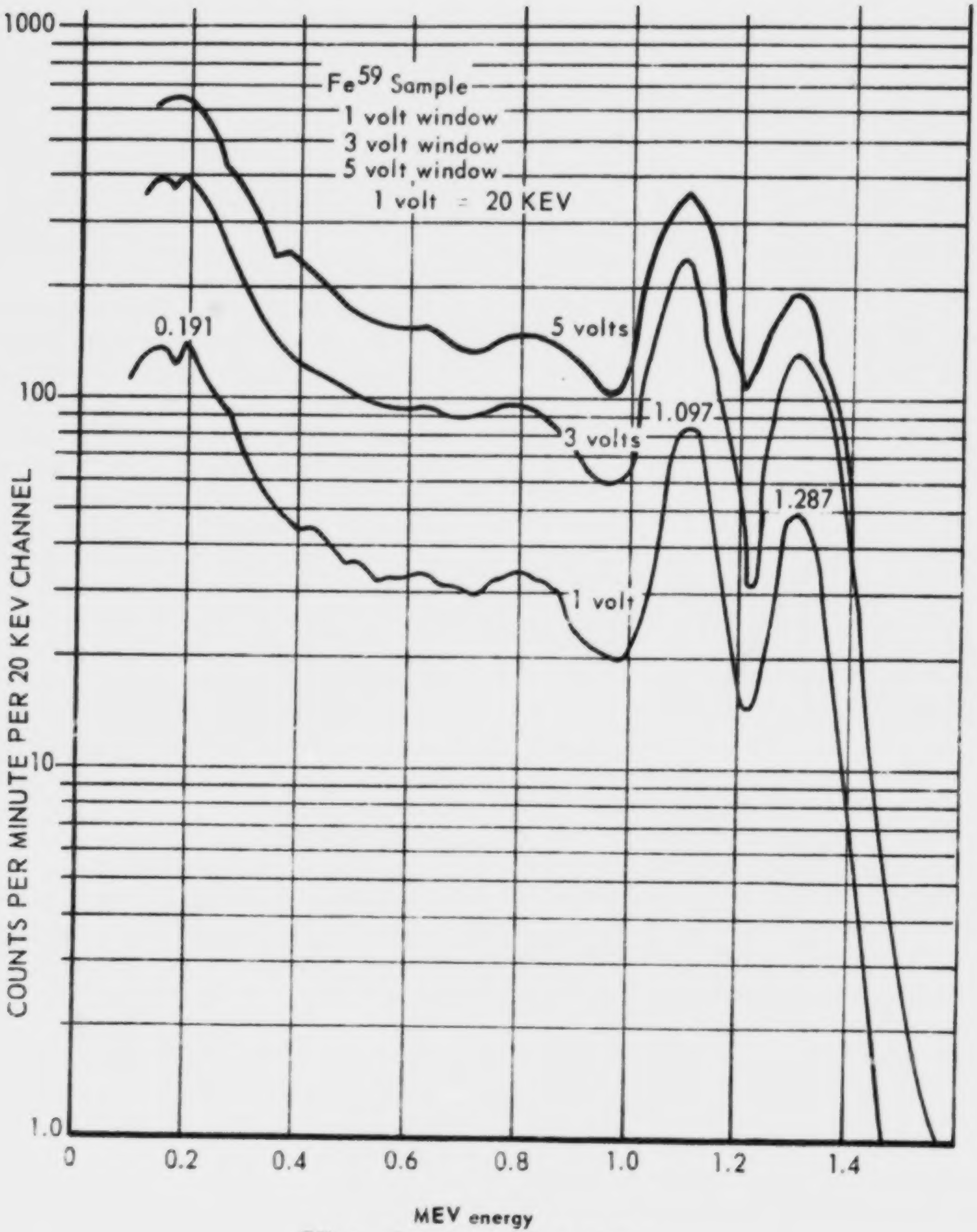
The single channel analyzer is ideally suited for counting single nuclides. One example is the counting of I^{131} separated from milk by ion-exchange resins.

The most critical parameter in single nuclide counting is the window width. To minimize the effects of instrument drift, a peak area should be chosen such that small changes in the baseline discriminator do not appreciably change the count rate. A second factor which must be considered is the effect of counting statistics.

The channel width is chosen to minimize the relative statistical error of the net sample count. If the channel width is too narrow, the gross (sample + background) counting rate ($N_{s + Bg}$) and the background counting rate (N_{Bg}) will be small. The net counting rate ($N_{s + Bg} - N_{Bg}$) will also be small and the relative error ν (equation 2)

$$\nu = \sqrt{\frac{\sigma^2}{X}} = \frac{\sqrt{\dfrac{N_{s + Bg}}{t_{s + Bg}} + \dfrac{N_{Bg}}{t_{Bg}}}}{N_{s + Bg} - N_{Bg}} \qquad (2)$$

will be large due to the poor statistics of small numbers. If the channel width is too wide, the gross (sample + background) counting rate and the background counting rate will be much larger than the net counting rate. The relative error of the net counting rate will again be large. The optimum channel width depends not only upon the ratio of the gross (sample plus background) to background counting rates but also upon the spread of the peak. Peak spread can be expressed in terms of full width at half peak height (F.W.H.P.H). The units may be pulse height units, volts, channels, or direct energy units such as Kev or Mev.



Effects of window width on spectra
Figure 3

The photopeak pulse height distribution is assumed to be a normal distribution, the background count rate is assumed to be a linear function of the pulse height, and the background is assumed to not displace the photopeak of the gross spectrum on the energy scale. Data for such a statistical evaluation has been developed by Reber and Major.(8) They have calculated the relative error in terms of Poisson counting statistics. The integral equation is differentiated in terms of the channel width and set equal to zero to find the minimum relative error. A plot of the data from such calculations is given in Figure 4.

The usefulness of the single-channel analyzer can be more fully realized by tracer studies using single nuclides and by using a gamma emitting isotope to determine the chemical yield of a chemical purification procedure. Examples of the latter include Ba^{133} for radium analysis and Sr^{85} for Sr^{90} analyses where Y^{90} is counted.

B. Simultaneous Equation Analysis

One technique for calculating the activity of a radionuclide from gamma spectral data is the method involving the solution of simultaneous equations (one equation for each gamma emitter present). An area of the spectra (usually several channels around a photopeak on a multi-channel analyzer) is chosen for each nuclide. This is an ideal application for the single-channel analyzer as a window can be set over in each peak area. In gamma spectral analysis of milk, the nuclides K^{40}, I^{131}, Cs^{137}, and $Ba\text{-}La^{140}$ are represented by four selected portions in the spectrum. Making four counts with the single-channel analyzer will accomplish the job.

There are several disadvantages or limitations in such a single-channel analyzer application, such as, no evidence of instrument drift or of the presence of other nuclides in the sample than those being analyzed for. In either case, considerable error could be introduced into the analysis. The application of the simultaneous equation technique to the single-channel analyzer can result in economies in instrumentation costs but with the greater possibility of unsuspected errors.

C. Criteria for Usefulness

The criteria of minimum detectable activity (M.D.A.) and minimum sensitivity (M.S.) are applicable to single channel analyses. They can be used to set the limit of detectability for both qualitative and quantitative analysis. The selection of operating conditions and in particular the window width will determine the M.D.A. and the M.S.

Another criteria that will aid in selection of counting conditions or making a choice between counting techniques is the Figure of Merit. (9) It is equal to the ratio of the number of samples that can be counted in a given time period with a given statistical accuracy for the two instruments or counting techniques being compared. The value of the figure of merit is independent of the statistical accuracy used as a criteria.

The figure of merit for comparing counting technique A to counting technique B or instrument A to instrument B is calculated by equation 2.

$$F(A/B) = \frac{(S_A)^2\ (S_B + 2B_B)}{(S_B)^2\ (S_A + 2B_A)} \qquad (3)$$

where

$F(A/B)$ is the figure of merit for instrument A compared to instrument B

S_A & S_B are the net sample count rates of the same standard source for instrument A and B respectively.

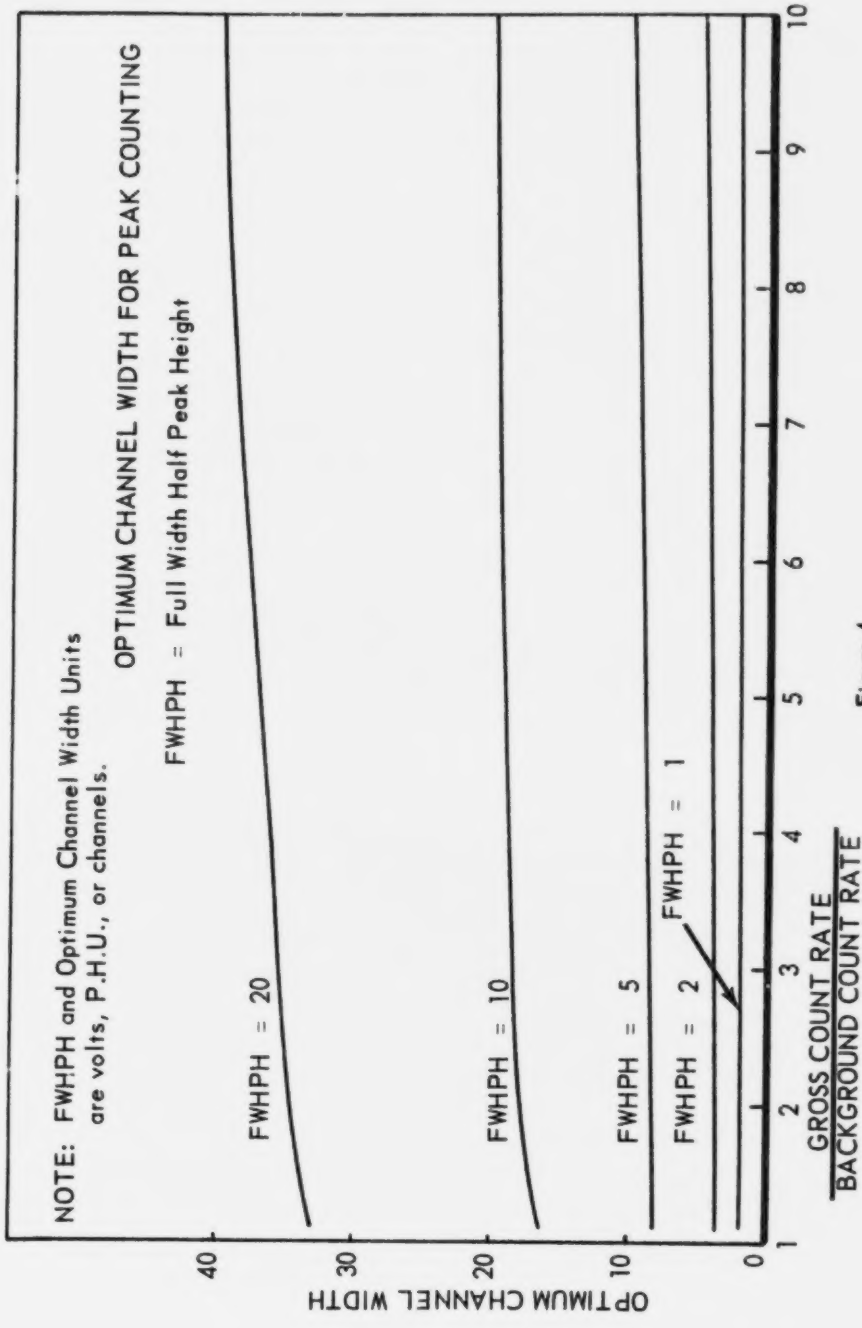

NOTE: FWHPH and Optimum Channel Width Units
are volts, P.H.U., or channels.
OPTIMUM CHANNEL WIDTH FOR PEAK COUNTING
FWHPH = Full Width Half Peak Height
FWHPH = 20
FWHPH = 10
FWHPH = 5
FWHPH = 2
FWHPH = 1
OPTIMUM CHANNEL WIDTH
40
30
20
10
0
1
2
3
4
5
6
7
8
9
10
GROSS COUNT RATE
BACKGROUND COUNT RATE

Figure 4

B_A & B_B are the background count rates for instruments A and B respectively.

With samples of high activity equation 3 reduces to equation 4.

$$F(A/B) = \frac{S_A}{S_B} \qquad (4)$$

While for samples with counting rates the order of background or less equation 3 reduces to equation 5.

$$F(A/B) = \frac{(S_A)^2 \; B_B}{(S_B)^2 \; B_A} \qquad (5)$$

It is evident that the figure of merit is dependent on the sample activity. This is more clearly seen by separating the sample count rate into its two components, sample disintegration rate and counting efficiency.

$$S_A = \text{dpm} \times E_A$$

$$S_B = \text{dpm} \times E_B$$

Where E_A and E_B are the counting efficiencies for instruments A and B respectively.

By substituting the components of sample count rate in equation 3 equation 6 is obtained.

$$F(A/B) = \frac{(\text{dpm}E_A)^2 (\text{dpm} \times E_B + 2\,B_B)}{(\text{dpm} \times E_B)^2 (\text{dpm} \times E_A + 2\,B_A)} \qquad (6)$$

Equation 6 can be reduced to equation 7.

$$\frac{E_A{}^2 (\text{dpm} \times E_B + 2\,B_B)}{E_B{}^2 (\text{dpm} \times E_A + 2\,B_A} \qquad (7)$$

It is desirable to count as many samples as possible in a given time period to a specific accuracy. Therefore, when F(A/B) is greater than 1, instrument A is superior to instrument B. If F(A/B) is 1, both instruments are equal; and if it is less than 1, instrument B is superior.

The instruments must be compared using a standard with activity close to that anticipated for actual sample counting. As the figure of merit is dependent on sample disintegration rate, it may be that one instrument is superior for one activity range while the other is superior for a different activity range.

These criteria are applicable to all instruments and counting techniques and will provide sound guidance in determining applications where the single-channel analyzer can be used to advantage. The figure of merit can also be used to determine whether differential or integral counting would be more appropriate. An example of this application for I^{131} counting follows:

Required: Determine the figure of merit for differential (A) versus integral (B) counting of I^{131} at activity levels of 1 to 100,000 dpm.

Given: Sample size = 100 ml in 250 ml bottle.

Differential Background for the 0.364 Mev peak (0.30 - 0.48 Mev) = 24 cpm

Differential Efficiency = 3.95%

Integral Background for the 0.364 Mev peak to ∞ = 127 cpm

Integral Efficiency = 4.5%

Solution: Reduce equation 7 so that dpm is the only variable.

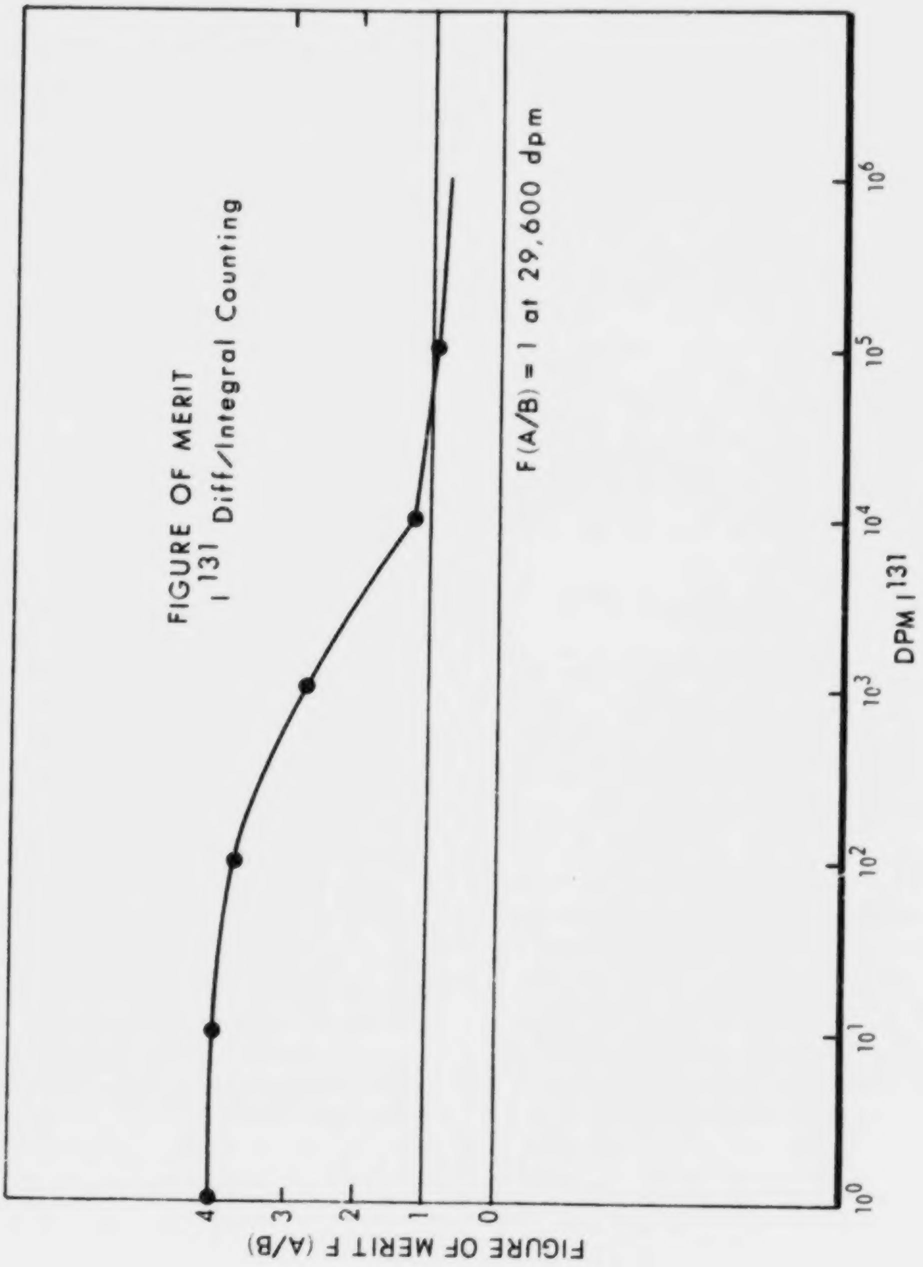


FIGURE OF MERIT
I^{131} Diff/Integral Counting
F(A/B) = 1 at 29,600 dpm
FIGURE OF MERIT F (A/B)
4
3
2
1
0
DPM I^{131}
10^0
10^1
10^2
10^3
10^4
10^5
10^6

Figure 5

$$F(A/B) = \frac{(dpm \times E_A)^2 \, (dpm \times E_B + 2\,B_B)}{(dpm \times E_B)^2 \, (dpm \times E_A + 2\,B_A)}$$

$$= \frac{(.0395 \text{ dpm})^2 \, (.045 \text{ dpm} + 254)}{(.045 \text{ dpm})^2 \, (.0395 \text{ dpm} + 48)}$$

$$= \frac{.00156 \, (.045 \text{ dpm} + 254)}{.00202 \, (.0395 \text{ dpm} + 48)}$$

$$= \frac{.00007 \text{ dpm} + .396}{.00008 \text{ dpm} + .097}$$

Substitute appropriate dpm values; 1, 10, 100, 1000, 10,000, and 100,000 dpm. The 10 dpm calculation is as follows:

$$\frac{.00007\,(10) + .396}{.00008\,(10) + .097} = \frac{.3967}{.0978} = 4.06$$

The data resulting from the calculations is plotted on Figure 5. The point where F(A/B) = 1 is seen to be at 29,600 dpm. Only at activity levels greater than this, would the integral counting setup be more efficient.

V. SUMMARY

There are a number of analytical applications where the single-channel gamma analyzer can be useful. These include identification and quantification of activities either after chemical separation or in complex mixtures. Tracer studies and carrier yield determinations are natural applications.

Versatility in application is determined by the capability of choosing appropriate instrumental operating conditions. Variable scan speeds, ratemeter time constants, and read-out speeds are prime examples of where flexibility is required.

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GPO 886-523

Matrix Algebra

INTRODUCTION

A matrix is a rectangular array of numbers enclosed by a pair of brackets which may or may not have a numerical value. Matrices can be added, subtracted, multiplied or divided if certain rules of operation are followed. The purpose of this discussion is to present these operations and to give certain definitions which will be helpful in solving problems envolving matrix algebra.

In the matrix

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} & \cdots & a_{1n} \\ a_{21} & a_{22} & a_{23} & \cdots & a_{2n} \\ a_{31} & a_{32} & a_{33} & \cdots & a_{3n} \\ . & . & . & \cdots & . \\ . & . & . & \cdots & . \\ a_{m1} & a_{m2} & a_{m3} & \cdots & a_{mn} \end{pmatrix}$$

the numbers or functions a_{ij} are called its elements. In the double subscript notation, the first subscript denotes the row and the second subscript denotes the column in which the element stands. Therefore, all elements in the second row have a 2 as the first subscript and all elements in the third column have a 3 as the second subscript. A matrix of m rows and n columns is defined as a "m by n" or "m × n" matrix. Instead of writing the matrix out in its entirety, it will be defined as "A". This notation will be used throughout this discussion.

II. ROW MATRIX

A row matrix is defined as a series of numbers arranged in a row

$$\text{"A"} = (a_{11} \quad a_{12} \quad a_{13} \quad \cdots \quad a_{1n})$$

III. COLUMN MATRIX

A column matrix is defined as a series of numbers arranged in a column.

$$\text{"A"} = \begin{pmatrix} a_{11} \\ a_{21} \\ a_{31} \\ a_{41} \\ . \\ . \\ a_{m1} \end{pmatrix}$$

IV. DIAGONAL MATRIX

A diagonal matrix is defined as a series of numbers arranged in a diagonal in a matrix. All other numbers making up the matrix are zero's. This means that all diagonal matrices are square, i.e., equal number of rows and columns, and only has numbers arranged in the main diagonal.

$$\text{"A"} = \begin{pmatrix} a_{11} & 0 & 0 & 0 \\ 0 & a_{22} & 0 & 0 \\ 0 & 0 & a_{23} & 0 \\ 0 & 0 & 0 & a_{44} \end{pmatrix}$$

EXAMPLE NO. 1

Arrange the row matrix "A" as a diagonal matrix

"A" = $(3\ 2\ 4\ 1)$ (row matrix)

"A" = $\begin{pmatrix} 3 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 4 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$ (diagonal matrix)

EXAMPLE NO. 2

Arrange the column matrix "A" as a diagonal matrix

"A" = $\begin{pmatrix} 1 \\ 2 \\ 3 \\ 4 \end{pmatrix}$ (column matrix)

"A" = $\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 3 & 0 \\ 0 & 0 & 0 & 4 \end{pmatrix}$ (diagonal matrix)

V. THE TRANSPOSE

The matrix of order "m × n" obtained by interchanging the rows and columns of an "m × n" matrix "A" is called the transpose and is denoted as "A^T".

If we have the matrix

"A" = $\begin{pmatrix} P_1 & P_2 & P_3 \\ P_4 & P_5 & P_6 \end{pmatrix}$

then the transpose of matrix "A" is

"A^T" = $\begin{pmatrix} P_1 & P_4 \\ P_2 & P_5 \\ P_3 & P_6 \end{pmatrix}$

EXAMPLE NO. 3

Write the transpose of matrix "A"

"A" = $\begin{pmatrix} 1 & 2 & 3 \\ 4 & 5 & 6 \\ 7 & 8 & 9 \end{pmatrix}$

"A^T" = $\begin{pmatrix} 1 & 4 & 7 \\ 2 & 5 & 8 \\ 3 & 6 & 9 \end{pmatrix}$

VI. MULTIPLICATION OF MATRIX

Matrices can be multiplied if they are multiplied in the order that an equation calls for. If the equation has the form "AB" where "A" and "B" are matrices then they must be multiplied in that order. Some simple rules which are helpful are

"AB" ≠ "BA" generally

"AB" = 0 does not necessarily imply "A" = 0 or "B" = 0

"AB" = "BC" does not necessarily imply "A" = "C"

The operation of multiplication is row by column; each element of the row is multiplied by the corresponding element of the column and then the products are summed. If the matrices "A" and "B" are given as

"A" = $(a_{11}\ a_{12}\ a_{13}\ a_{14})$ "B" = $\begin{pmatrix} b_{11} \\ b_{21} \\ b_{31} \\ b_{41} \end{pmatrix}$

then the product "AB" = "C" is given as

"C" = $(a_{11}b_{11} + a_{12}b_{21} + a_{13}b_{31} + a_{14}b_{41})$

EXAMPLE NO. 4

Find the product of matrices "A" and "B".

$$\text{"A"} = (1 \quad 2 \quad 3) \qquad \text{"B"} = \begin{pmatrix} 4 \\ 5 \\ 6 \end{pmatrix}$$

then

$$\text{"AB"} = \text{"C"} = \left[1(4) + 2(5) + 3(6)\right] = \left[32\right]$$

The operation of multiplication of higher order matrices is again row by column. Each element in the first row (matrix "A") is multiplied by the corresponding element in the first column (matrix "B") and their products summed. Then each element in the first row (matrix "A") is multiplied by the corresponding element in the second column (matrix "B") and their products summed. This will give the first two row elements in the product matrix "C". This process is continued until all rows (matrix "A") have been multiplied by all columns (matrix "B"). Therefore, by definition, to multiply two matrices the number of elements in the rows (matrix "A") and the number of elements in the columns (matrix "B") must be equal.

If two matrices "A" and "B" are given as

$$\text{"A"} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \\ a_{31} & a_{32} \end{pmatrix} \qquad \text{"B"} = \begin{pmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \end{pmatrix}$$

then "AB" = "C" where

$$\text{"C"} = \begin{pmatrix} a_{11}b_{11} + a_{12}b_{21} & a_{11}b_{12} + a_{12}b_{22} \\ a_{21}b_{11} + a_{22}b_{21} & a_{21}b_{12} + a_{22}b_{22} \\ a_{31}b_{11} + a_{32}b_{21} & a_{31}b_{12} + a_{32}b_{22} \end{pmatrix}$$

EXAMPLE NO. 5

Find the product of matrices "A" and "B"

$$\text{"A"} = \begin{pmatrix} 2 & 3 \\ 1 & 5 \\ 4 & 6 \end{pmatrix} \qquad \text{"B"} = \begin{pmatrix} 1 & 2 \\ 4 & 0 \end{pmatrix}$$

$$\text{"AB"} = \text{"C"} = \begin{pmatrix} 2(1) + 3(4) & 2(2) + 3(0) \\ 1(1) + 5(4) & 1(2) + 5(0) \\ 4(1) + 6(4) & 4(2) + 6(0) \end{pmatrix}$$

$$\text{"C"} = \begin{pmatrix} 14 & 4 \\ 21 & 2 \\ 28 & 8 \end{pmatrix}$$

EXAMPLE NO. 6

Find the product of matrices "A" and "B"

$$\text{"A"} = (1 \quad 2 \quad 3) \qquad \text{"B"} = \begin{pmatrix} 4 & -6 & 9 & 6 \\ 0 & -7 & 10 & 7 \\ 5 & 8 & -11 & -8 \end{pmatrix}$$

$$\text{"AB"} = \text{"C"} = [1(4) + 2(0) + 3(5) \quad 1(-6) + 2(-7)$$

$$+ 3(8) \quad 1(9) + 2(10) + 3(-11)$$

$$1(6) + 2(7) + 3(-8)]$$

$$\text{"C"} = (19 \quad 4 \quad -4 \quad -4)$$

EXAMPLE NO. 7

Find the product of matrices "A" and "B"

$$\text{"A"} = \begin{pmatrix} 2 & 3 & 4 \\ 1 & 5 & 6 \end{pmatrix} \qquad \text{"B"} = \begin{pmatrix} 1 \\ 2 \\ 3 \end{pmatrix}$$

$$\text{"AB"} = \text{"C"} = \begin{pmatrix} 2(1) + 3(2) + 4(3) \\ 1(1) + 5(2) + 6(3) \end{pmatrix}$$

$$\text{"C"} = \begin{pmatrix} 20 \\ 29 \end{pmatrix}$$

VII. THE INVERSE

When dividing matrices the denominator can be written as an inverse and then the two matrices can be multiplied. If the matrix "A" is to be divided by the matrix "B".

$$\frac{\text{"A"}}{\text{"B"}} = \text{"C"}$$

this can be rewritten as

$$\text{"AB}^{-1}\text{"} = \text{"C"}$$

where "B^{-1}" is the inverse of "B"

The inverse is defined as

$$\text{"B}^{-1}\text{"} = \frac{\text{adjoint "B"}}{|\text{"B"}|}$$

where "B" is the evaluated numerical value of matrix "B" and the adjoint "B" is defined as follows. If "B" is a matrix

$$\text{"B"} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix}$$

then the adjoint (adj) is found by expanding in the following manner. If the element in the first row and first column is considered, (a_{11}), that row and column that the element is contained in, are crossed out leaving the element (a_{22}). This becomes the first element in the first row and first column of the matrix adj "B". If the second element in the first row is considered, (a_{12}), that row and column that the element is contained in, are crossed out leaving the term (a_{21}). This becomes the element in the first column and second row of the adj (B) matrix. It has a negative sign attached because of its position. The signs alternate, positive and negative, going across a row or down a column. If the first element in the second row is considered, (a_{21}), that row and column that the element is contained in, are crossed out leaving the term (a_{12}). This becomes the element in the second column and first row of the adj (B) matrix. It has a negative sign attached because of its position. If the second element in the second column is considered, (a_{22}), that row and column that the element is contained in are crossed out leaving the term (a_{11}). This becomes the element and the second row and second column of the adj B matrix. The adj B matrix then becomes

$$\text{Adj "B"} = \begin{pmatrix} a_{22} & -a_{12} \\ -a_{21} & a_{11} \end{pmatrix}$$

EXAMPLE NO. 8

Find the inverse of matrix "A"

$$\text{"A"} = \begin{pmatrix} 2 & 3 \\ 1 & 4 \end{pmatrix}$$

$$|\text{"A"}| = \left[2(4) - 1(3)\right] = 5$$

$$\text{Adj "A"} = \begin{pmatrix} 4 & -3 \\ -1 & 2 \end{pmatrix}$$

then by definition

$$\text{"A}^{-1}\text{"} = \frac{\text{adj "A"}}{|\text{"A"}|} \quad \frac{\begin{pmatrix} 4 & -3 \\ -1 & 2 \end{pmatrix}}{5}$$

$$\text{"A}^{-1}\text{"} = \begin{pmatrix} 4/5 & -3/5 \\ -1/5 & 2/5 \end{pmatrix}$$

If a higher order matrix B is

$$"B" = \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix}$$

the adj of "B" is found by expanding in the following manner. If the element in the first row and first column is considered (a_{11}), that row and column that the element is contained in, are crossed out leaving a sub-matrix

$$\begin{pmatrix} a_{22} & a_{23} \\ a_{32} & a_{33} \end{pmatrix}$$

this sub-matrix becomes the element in the first row and first column of the matrix adj B. If the second element in the first row is considered, a_{12}, that row and column that the element is contained, are crossed out leaving a sub-matrix

$$\begin{pmatrix} a_{21} & a_{23} \\ a_{31} & a_{33} \end{pmatrix}$$

this sub-matrix becomes the element in the second row and first column. This sub-matrix has a negative sign attached because of its position. If the third element in the first row is considered, a_{13}, that row and column, that the element is contained in are crossed out leaving a sub-matrix

$$\begin{pmatrix} a_{21} & a_{22} \\ a_{31} & a_{32} \end{pmatrix}$$

this sub-matrix becomes the element in the third row and first column. It has a positive sign because of its position. If the first element in the second row is considered, a_{21}, that row and column that the element is contained in, are crossed out leaving a sub-matrix

$$\begin{pmatrix} a_{12} & a_{13} \\ a_{32} & a_{33} \end{pmatrix}$$

this sub-matrix becomes the element in the first row and second column. It has a negative sign attached because of its position. This process is carried out until nine sub-matrices have been formed and are so arranged.

$$\text{Adj } "B" = \begin{pmatrix} \begin{pmatrix} a_{22} & a_{23} \\ a_{32} & a_{33} \end{pmatrix} & -\begin{pmatrix} a_{12} & a_{13} \\ a_{32} & a_{33} \end{pmatrix} & \begin{pmatrix} a_{12} & a_{13} \\ a_{22} & a_{23} \end{pmatrix} \\ -\begin{pmatrix} a_{21} & a_{23} \\ a_{31} & a_{33} \end{pmatrix} & \begin{pmatrix} a_{11} & a_{13} \\ a_{31} & a_{33} \end{pmatrix} & -\begin{pmatrix} a_{11} & a_{13} \\ a_{21} & a_{23} \end{pmatrix} \\ \begin{pmatrix} a_{21} & a_{22} \\ a_{31} & a_{32} \end{pmatrix} & -\begin{pmatrix} a_{11} & a_{12} \\ a_{31} & a_{32} \end{pmatrix} & \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \end{pmatrix}$$

The individual sub-matrices can be evaluated numerically to give

$$\text{adj "B"} = \begin{pmatrix} C_{11} & C_{12} & C_{13} \\ C_{21} & C_{22} & C_{23} \\ C_{31} & C_{32} & C_{33} \end{pmatrix}$$

where

$$\text{"}C_{11}\text{"} = \begin{pmatrix} a_{22} & a_{23} \\ a_{32} & a_{33} \end{pmatrix}$$

$$\text{"}C_{11}\text{"} = (a_{22}a_{33} - a_{32}\,a_{23})$$

$$\text{"}C_{21}\text{"} = -\begin{pmatrix} a_{21} & a_{23} \\ a_{31} & a_{33} \end{pmatrix}$$

$$\text{"}C_{21}\text{"} = -(a_{21}a_{33} - a_{31}a_{23})$$

The numerical value, $|\text{"B"}|$, is found by ordinary determinate methods.

EXAMPLE NO. 9

Find the inverse of the matrix "A"

$$\text{"A"} = \begin{pmatrix} 2 & 3 & 1 \\ 1 & 2 & 3 \\ 3 & 1 & 2 \end{pmatrix}$$

$$|\text{"A"}| = 2(2)\,(2) + 3(3)\,(3) + 1(1)\,(1) - 3(2)\,(1)$$

$$- 1(3)\,(2) - 1(3)\,(2)$$

$$= 8 + 27 + 1 - 6 - 6 - 6$$

$$= 18$$

$$\text{Adj "A"} = \begin{pmatrix} \begin{pmatrix} 2 & 3 \\ 1 & 2 \end{pmatrix} & -\begin{pmatrix} 3 & 1 \\ 1 & 2 \end{pmatrix} & \begin{pmatrix} 3 & 1 \\ 2 & 3 \end{pmatrix} \\ -\begin{pmatrix} 1 & 3 \\ 3 & 2 \end{pmatrix} & \begin{pmatrix} 2 & 1 \\ 3 & 2 \end{pmatrix} & -\begin{pmatrix} 2 & 1 \\ 1 & 3 \end{pmatrix} \\ \begin{pmatrix} 1 & 2 \\ 3 & 1 \end{pmatrix} & -\begin{pmatrix} 2 & 3 \\ 3 & 1 \end{pmatrix} & \begin{pmatrix} 2 & 3 \\ 1 & 2 \end{pmatrix} \end{pmatrix}$$

which upon evaluating numerically the individual sub-matrices

$$\text{Adj "A"} = \begin{pmatrix} 1 & -5 & 7 \\ 7 & 1 & -5 \\ -5 & 7 & 1 \end{pmatrix}$$

Then by definition

$$\text{"A-1"} = \frac{\text{adj "A"}}{|\text{"A"}|}$$

$$\text{"A-1"} = \frac{\begin{pmatrix} 1 & -5 & 7 \\ 7 & 1 & -5 \\ -5 & 7 & 1 \end{pmatrix}}{18}$$

$$\text{"A-1"} = \begin{pmatrix} 1/18 & -5/18 & 7/18 \\ 7/18 & 1/18 & -5/18 \\ -5/18 & 7/18 & 1/18 \end{pmatrix}$$

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GPO 884-555

Least Squares Analysis of Gamma Spectra

I. INTRODUCTION

Samples which are to be analyzed quantitatively, by gamma spectrometry can be handled in many ways: Compton continuum subtraction, spectrum stripping, simultaneous equations, etc. A relatively new method, has been developed, which is in many ways, superior to these standard procedures.

It is known that there are counting errors associated with the gross count rate obtained from counting a sample. Errors are also introduced when the total spectra is analyzed to obtain levels of activity of specific radionuclides.

The "Least Squares" method is used for obtaining the best estimates, of the amounts of each radionuclide present in a sample and an evaluation of the error of these results.

II. LEAST SQUARES

The least squares method is a mathematical treatment of data where a "best fit" curve is mathematically computed such that the sum of the squares of the deviations from the "best fit" curve is a minimum. The method by which this curve is fitted rests upon the assumption that the scatter of the individual observations around the fitted line will approximate a normal distribution. It also states that the observations should have a constant variance (square of the standard deviation) and the mean of their errors should be zero. Since radioactive decay is a random process, the assumptions are satisfied except that the variance will not be constant. This is corrected by use of a weighted element in the procedure.

In view of the preceding discussion, an example is now worked to illustrate the use of the least squares method. Hypothetical data[1] has been collected, Table 1, and is plotted in Figure 1.

EXAMPLE NO. 1

Table 1

	Observed y	Observed x	x^2	xy	Calculated y
	44	5.3	28.1	233	39.0
	20	3.5	12.3	70.0	27.6
	24	3.0	9.0	72.0	24.5
	14	1.2	1.44	16.8	13.2
	12	0.48	0.23	5.8	8.6
	3	-0.26	0.07	-0.8	4.0
Sums	117	13.22	51.14	396.8	116.9
Means	19.5	2.20	8.52	66.1	19.5

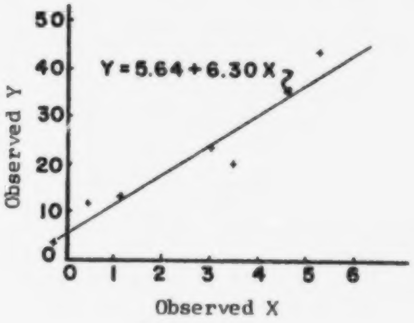

Figure 1

The normal equations, derived from calculus, yield

$$a + b\left(\frac{\Sigma x}{n}\right) - \left(\frac{\Sigma y}{n}\right) = 0 \qquad (1)$$

$$a\left(\frac{\Sigma x}{n}\right) + b\left(\frac{\Sigma x^2}{n}\right) - \left(\frac{\Sigma xy}{n}\right) = 0 \qquad (2)$$

where

n = number of observations = 6

$a + 2.20\,b - 19.5 = 0$

$2.20\,a + 8.52\,b - 66.1 = 0$

Solving for the constants "a" "b" gives

$$a = 5.64$$

$$b = 6.30$$

This gives the final equation,

$$y = 5.64 + 6.30\,x$$

from which the calculated values of "y" are obtained. This represents the best fit line through the scattered points, Figure 1.

This principle of least squares curve fitting is applied to gamma spectroscopy in the following way. Assume that a background has been taken and that for illustrative purposes the first five channels are the channels of interest.

The data is graphically plotted in Figure 2. Because background represents a random process, there will be an error associated with each individual channel count. In order to find the most accurate value in each channel a mathematical analysis must be performed. An extension of least squares method is used to determine a curve such that the deviations from this curve are smaller than the deviations from any other curve drawn through these points. At the same time the equation of the curve of best fit is calculated. This means that the calculated curve is not the "true" curve but it does have some error associated with it, however, it does represent the best estimate of the "true" curve.

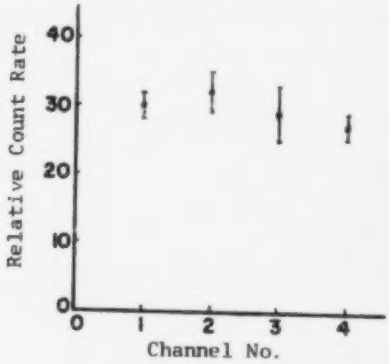

Figure 2

This principle can be applied to composite spectra. Looking at Figure 3 it is seen that one radionuclide plus a background are plotted.

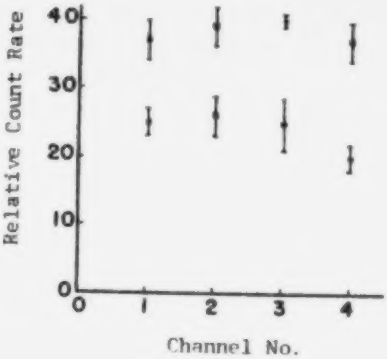

Figure 3

To understand how this is plotted, observe one channel, Figure 4.

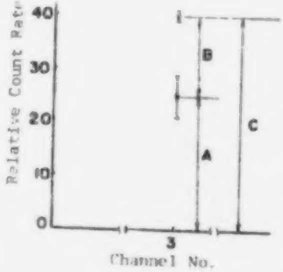

Figure 4

"A" is the relative count rate due to background which has an associated error. "B" is the relative count rate due to nuclide X and it also has an error associated with it. Therefore "C" is the gross relative count rate in channel 3. In this gross count rate there are the two associated errors due to "A" and "B" combined into one error that is associated with the gross count rate, "C". The problem in quantitative analysis is to breakdown the gross count rate "C" into a background count rate and a count rate due to nuclide X. This separation must be done in such a way that the errors associated with each component and the actual value obtained for each component are the best statistical answers. Again, the least squares method is used to obtain this best statistical answer. The separation is accomplished by writing an equation for each channel which has a term for the nuclide, the background and an error term for each. By writing this series of equations in either summation or matrix notation they are relatively easy to solve. An example might best show this reasoning.

EXAMPLE NO. 2

Assume that an equation, written in summation notation, has been set up of the following form:

$$y = \sum_{i=1}^{n} x_{ij} \quad B_j \pm \epsilon_i \qquad (3)$$

where

i = channel number ($n = 100$)

y = gross count rate in channel i

j = nuclide (designation assigned)

B_j = disintegration rate of the j^{th} nuclide

x_{ij} = ratio of the count rate to the disintegration rate for the standard of the j^{th} nuclide in channel i

ϵ_i = random error in channel i

Assume now that a standard of Cs^{137} has been counted for one minute and that 1000 dpm of Cs^{137} gave a net count rate of 10 cpm in channel 33. Our error term associated with this is $\sqrt{10} = 3.3$ cpm. Now an unknown amount of Cs^{137} is counted for one minute and gives 20 cpm in channel 33. Substituting into the equation yields

$$20 = \frac{10}{1000} \; B_{cs} \pm \sqrt{20}$$

$$20 = 0.01 \; B_{cs} \pm 4.2$$

$$20 \pm 4.2 = 0.01 \; B_{cs}$$

$$B_{cs} = \frac{20 \pm 4.2}{0.01} = 2000 \pm 420 \text{ dpm}$$

Without the error term a value of 2000 dpm would have been calculated which is the answer if the same problem had been worked by the simultaneous equation method.

Although the example just worked is relatively simple, more complex problems can be worked using the same general idea. The big advantage of the method of least squares lies in the fact that all channels (usually 100) are used and that an error can also be calculated for each channel. Also the activity (dpm) is calculated from each channel instead of selected channels.

Other advantages of this method are:(2)

1. The methód is free from subjective errors, i.e., personal bias.

2. It is amendable to computer calculation. In some of the more complex spectra analysis it is almost impossible to do the calculations by hand.

3. It should be particularly suited to spectra whose statistical variations in counting are large. Because radioactive decay is a statistical process this method is well suited for this analysis.

4. It should be possible to analyze spectra where peaks are super-imposed. This is a particularly good advantage as additional analysis (spectrum stripping) will not have to be applied before a quantitative analysis method can be employed.

5. Estimates may be made of the variance, and the uncertainty of the variance associated with each result.

6. Spectra which contain nuclides not accounted for in the original library of standards, can be analyzed by applying the appropriate correction factors.

The method is not without disadvantages. Some of these are:(2)

1. The number of nuclides has to be less than or equal to the number of channels observed. In most cases this will not even enter into the scope.

2. The necessity of applying smoothing corrections to nullify the effect of small inaccuracies and statistical counting fluctuations in an ill-conditioned matrix.

3. The statistical counting fluctuations of the standards must be negligible in comparison with the sample. Therefore the standards have to be counted for a very long time.

4. Have to gather sufficient counts so no channel has zero counts in the same spectra.

5. Because of the sensitivity of the method, very good instrument stability must be maintained. The method analyzes each individual channel instead of selected channels so drift must be minimized and calibration must be precise.

Now that some of the advantages and disadvantages of the method have been mentioned, the mathematical model derived from least squares will be explained.(2)

The gamma spectrometer is considered to have "n" number of channels numbered 1 . . . i . . . n and the midpoint of each channel represents a gamma energy E_i. The observed count rate for each value of E_i can be found experimentally. For m nuclides this results in an "m x n" matrix A which is defined as our standard library, Table 2. The background can be considered as another nuclide in the standard library.

Table 2. STANDARDS LIBRARY

Channel No.	Nuclide X $\frac{cpm}{dpm}$	Nuclide Y $\frac{cpm}{dpm}$	Nuclide Z $\frac{cpm}{dpm}$
1	4	2	1
2	3	6	5
3	2	7	4
4	1	5	3
5	0	4	1

From Table 1 our "A" matrix is written as

$$A = \begin{pmatrix} 4 & 2 & 1 \\ 3 & 6 & 5 \\ 2 & 7 & 4 \\ 1 & 5 & 3 \\ 0 & 4 & 1 \end{pmatrix}$$

A sample spectrum, p, which is the result of a mixture of "m" nuclides ($m \leq n$) each corresponding to the energies E_i, results in a set of linear simultaneous equations, written in matrix notation,

$$A \beta = p \qquad (4)$$

where

A = the standard library matrix as defined above $(\frac{cpm}{dpm})$

β = unknown intensity of each nuclide in dpm

p = observed sample spectra (cpm)

Solving for β yields

$$\beta = A^{-1} p \qquad (5)$$

It is assumed that all nuclides in the standard library have been counted long enough so that the only errors encountered are statistical fluctuation in "p", our sample spectra. For this reason the principle of least squares can be applied. By minimizing the sum of the squares of errors in "p", it can be shown from the theory of multi-linear regression that the resulting normal equations derived are

$$A^T A \beta = A^T p \qquad (6)$$

Another principle of least squares requires that the variance of each observation shall be a constant. Since the variance of each element in "p" is not constant, a diagonal weighting matrix "w" must be introduced where "w" = $1/\sigma^2$. In the simplest case $\sigma i^2 = p_i$, if the counting time in each channel is constant and there is no variance in our standard library.

The resulting equation is

$$A^T w A \beta = A^T w p \qquad (7)$$

Solving for the unknown β yields

$$\beta = (A^T w A)^{-1} \quad A^T w p \qquad (8)$$

After the β's have been determined, it is relatively easy to calculate the error associated with each β value. Through further derivation it can be shown that

$$\sigma^2(\beta_i) = C_{ii}^{-1}$$

which says that the standard deviation squared of any value of β is equal to the corresponding element in the inverse matrix C^{-1},

where

$$C^{-1} = (A^T w A)^{-1}$$

A simple example is now worked to show the mechanics of the method.

Channel No. i	Standard Library cpm/dpm			Measured Spectra p cpm
	Nuclide X	Nuclide Y	Nuclide Z	
1	1	1	1	3
2	1	1	0	2
3	1	0	0	3

Our "A" matrix, from our Standard Library is

$$A = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}$$

$$A^T = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}$$

$$p = \begin{pmatrix} 3 \\ 2 \\ 3 \end{pmatrix}$$

Our weighting diagonal matrix is

$$w = \begin{pmatrix} 1/3 & 0 & 0 \\ 0 & 1/2 & 0 \\ 0 & 0 & 1/3 \end{pmatrix}$$

Using our basic equation

$$\beta = (A^T wA)^{-1} A^T wp$$

We solve for β

$$(A^T w) = \begin{pmatrix} 1/3 & 1/2 & 1/3 \\ 1/3 & 1/2 & 0 \\ 1/3 & 0 & 0 \end{pmatrix}$$

$$(A^T wA) = \begin{pmatrix} 7/6 & 5/6 & 1/3 \\ 5/6 & 5/6 & 1/3 \\ 1/3 & 1/3 & 1/3 \end{pmatrix}$$

$$(A^T wA)^{-1} = \begin{pmatrix} 3 & -3 & 0 \\ -3 & 5 & -2 \\ 0 & -2 & 5 \end{pmatrix}$$

$$(A^T wA)^{-1} A^T = \begin{pmatrix} 0 & 0 & 3 \\ 0 & 2 & -3 \\ 3 & -2 & 0 \end{pmatrix}$$

$$(A^T wA)^{-1} A^T w = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & -1 \\ 1 & -1 & 0 \end{pmatrix}$$

$$(A^T wA)^{-1} A^T wp = \begin{pmatrix} 3 \\ -1 \\ 1 \end{pmatrix} = \beta$$

This means that nuclide X has 3 dpm, nuclide Y has -1 dpm and nuclide Z has 1 dpm from one measured spectra. If it is known that the intensities must be positive, i.e., $\beta_i \geq 0$, then by some method the negative element must be eliminated. One must be careful in eliminating negative elements because of the restrictions placed on the method. The negative value for β is indicative that the nuclide is not present. Therefore, that nuclide is eliminated from the standard library and equation 8 is recalculated.

Upon eliminating the negative element our new "A" matrix becomes

$$A = \begin{pmatrix} 1 & 1 \\ 1 & 0 \\ 1 & 0 \end{pmatrix}$$

$$A^T = \begin{pmatrix} 1 & 1 & 1 \\ 1 & 0 & 0 \end{pmatrix}$$

$$w = \begin{pmatrix} 1/3 & 0 & 0 \\ 0 & 1/2 & 0 \\ 0 & 0 & 1/3 \end{pmatrix}$$

Now re-evaluating the basic equation

$$\beta = (A^T wA)^{-1} A^T wp$$

$$(A^T w) = \begin{pmatrix} 1/3 & 1/2 & 1/3 \\ 1/3 & 0 & 0 \end{pmatrix}$$

$$(A^T wA) = \begin{pmatrix} 7/6 & 1/3 \\ 1/3 & 1/3 \end{pmatrix}$$

$$(A^T wA)^{-1} = \begin{pmatrix} 6/5 & -6/5 \\ -6/5 & 21/5 \end{pmatrix}$$

$$(A^T wA)^{-1} A^T w = \begin{pmatrix} 0 & 3/5 & 2/5 \\ 1 & -3/5 & -2/5 \end{pmatrix}$$

$$(A^T wA)^{-1} A^T wp = \begin{pmatrix} 12/5 \\ 3/5 \end{pmatrix} = \beta$$

To calculate the error associated with each value of the β's

$$\sigma^2(\beta_i) = C^{-1}_{ii} \quad \text{where } C^{-1} = (A^T wA)^{-1}$$

Therefore

$$\sigma^2(\beta_1) = 6/5$$
$$\sigma = \sqrt{1.2}$$
$$\sigma = 1.095$$
$$\sigma^2(\beta_3) = 21/5$$
$$\sigma = \sqrt{4.2}$$
$$\sigma = 2.05$$

The 95% confidence intervals for β_1 and β_3 expressed in dpm are

$$\beta_1 = 2.4 \pm 2.19$$
$$\beta_3 = 0.6 \pm 4.1$$

Although these error terms seem high it is due to the makeup of the problem. In general the accuracy of the method is on the order of 0.5 to 2% with greater accuracy obtainable with higher activity samples.

III. SUMMARY

Least square analysis of gamma-ray spectra is another method used in quantitative analyses. It has the advantage over other methods of being able to analyze all channels instead of the usual selected channels, being able to compute the variance of the answers and to analyze spectra where peaks are superimposed. Although the method is long and involved, computational wise, it lends itself to computer application very well.

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Evaluation of Quantitative Analysis Techniques

I. INTRODUCTION

In general there are two basic techniques by which an environmental sample may be analyzed utilizing gamma spectroscopy. First, the sample may be subjected to radiochemical separation and then the individual portions counted independently. Second, the sample may be counted in toto and the resulting complex spectrum analyzed by one of several spectral analysis techniques. These techniques are: Compton Continuum Subtraction, Spectrum Stripping, Simultaneous Equations, and Least Squares Analysis. A combination of radiochemical separation and spectral analysis may also be applied.

A question arises as to the comparative reliability and limits of detectability of these different procedures and where they may be used most effectively. The following is a discussion of the relative advantages and disadvantages and the precision of these procedures.

II. GENERAL CONSIDERATIONS

It should be pointed out at this point that it is difficult to establish absolute, realistic values for the analytical errors associated with the various analysis techniques. All measurements of concentration have associated with them inherent variations and inaccuracies. These errors are not only dependent on the method of chemical or spectral analysis but also on the sample counting rate and counting time, the amount of interference from other nuclides, the background count, and the media being analyzed. For purposes of comparison precision and minimum detectability of the different procedures will be presented in reference to the analysis of milk samples since this is where gamma spectroscopy is being utilized most extensively. The values presented may be assumed to generally hold true for the analysis of any other media.

Precision is given in terms of the $\pm 2\sigma$ range about the measured concentration corresponding to a 95% certainty that the true concentration is within that range. The minimum detectability is defined as that activity which results in a 100% error at the 95% confidence level for a 50 minute count.

III. CHEMICAL SEPARATION

A. General

Chemical separation is most often used as a qualitative tool in the analysis of complex gamma spectra. It can, however, be used quantitatively. A sample may be broken down completely and the individual nuclides counted on a single channel analyzer, or it may be partially simplified and counted on a multichannel analyzer with subsequent spectral analysis.

B. Evaluation

1. ADVANTAGES

a. The main advantage of chemical separation is its usefulness as a qualitative check on the makeup of the sample.

b. If a multi-channel analyzer is not available it is often the only practical method of analysis.

c. Where only one radionuclide is being analyzed, chemical separation may be the quickest and most accurate procedure.

2. DISADVANTAGES

a. It is often time consuming.

b. Introduces the possibility of error resulting from sample handling and processing.

3. MINIMUM DETECTABILITY

In general a minimum detectability of about 5-10 pc/l can be expected for a normal chemical separation.

4. PRECISION

It is difficult to give an absolute value for the precision of a radiochemical separation in general since it depends on the procedure and technique used. As a ballpark figure, precision for individual determinations within ± 10% may be expected depending on carrier yield. To this then must be added the error associated with the analysis and counting of the separated portions which is primarily dependent on the activity and counting time. For activities equal or greater than 100 pc/l, a total error within ±10% can be expected. For concentrations as low as 10 pc/l, an error of as much as ±50% can be expected.

5. USES

The primary uses of this procedure are: as a spot check on the radionuclide content of samples, where only a single channel analyzer is available, or where only one radionuclide is being analyzed for.

IV. SPECTRAL ANALYSIS

Spectral analysis techniques possess general two basic advantages over radiochemical techniques in the analysis of complex environmental samples. First, they have the advantage of a saving in time and labor once the procedure is established. Second, many errors resulting from sample handling and preparation are eliminated.

A. Compton Continuum Subtraction

1. GENERAL

The objective of this procedure is to eliminate by approximation the contribution to a given photopeak from interferring radionuclides. It is only an approximate graphical solution and should be evaluated as such.

2. EVALUATION

a. Advantages

The only real advantage to this procedure is that it is simple and quick.

b. Disadvantages

1. As already indicated this technique yields only an approximate quantitative evaluation.

2. Separate peaks for each radionuclide being analyzed must be present--the method cannot quantitatively separate overlapping peaks.

3. The counting efficiency must be determined by the same approximate procedure, i.e., by graphically subtracting out the compton contribution to the photopeak of interest in the standard spectrum. This introduces another source of error.

c. Minimum detectability

As previously indicated, the minimum detectability is dependent on the counting time and background. For a counting time of about 50 minutes, a minimum detectability of between 10-30 pc/ may be obtained.

d. Precision

This method cannot be expected to yield results with any better than ± 10% error. Between ±10 and ±30% error is probably a realistic range.

e. Uses

This procedure can be used on a day to day, routine basis but only as a gross indicator. If accurate concentrations are required, one of the other procedures must be used.

B. Spectrum Stripping

1. GENERAL

This is a procedure which is applicable to both quantitative and qualitative analysis. It may be used independently or in conjunction with other procedures, depending on the complexity of the spectrum.

2. EVALUATION

a. Advantages

1. The contribution of "hidden" peaks may be determined.

2. Automatic spectrum strippers can make the procedure useful on a day to day routine.

3. The procedure is applicable to computers.

b. Disadvantages

1. This procedure concentrates subtraction errors in the nuclides of lowest energy. Consequently evaluation of low energy gammas is less accurate than for high energy gammas when both are simultaneously present.

2. It is not possible to make measurements on a particular peak until all other peaks of higher energy have been stripped out.

3. Sum peaks are not accounted for.

4. Overlapping peaks cannot be separated except by trial and error.

5. The procedure is tedious and time consuming unless handled in an automatic spectrum stripper or computer.

c. Minimum detectability

For count rates of about 50 minutes a minimum detectability of about 10 pc/l can be expected.

d. Precision

It is difficult to evaluate this procedure as to precision since it depends so much on instrument characteristics (stability, etc.). In general, however, results with a ± 10% error for activities of 100 pc/l should be expected, with more accurate results in the higher energy range.

e. Uses

This procedure is not used generally on a routine basis unless an automatic spectrum stripping device is incorporated. It is useful as a spot check for any new radionuclides or as a check on the other analysis techniques.

C. Simultaneous Equation Method

1. GENERAL

This procedure entails a mathematical approach for the elimination of Compton interference from a given photopeak.

2. EVALUATION

a. Advantages

1. As long as not too many radionuclides are being analyzed, and the composition of the sample is constant day to day, the method provides an easy means of quantitative analysis.

2. The procedure is particularly applicable to computer handling.

b. Disadvantages

1. Radionuclides other than those accounted for in the equation cannot be present if the solution is to be valid.

2. A different photopeak must be present for each radionuclide. Partially overlapping peaks can be analyzed however.

3. A computer must be used to develop the equation if a significant number of nuclides are present.

c. Minimum detectability

The minimum detectability of this procedure is about 10 pc/l.

d. Precision

The precision is about ±5-10% for concentrations >100 pc/l and about ±5-10 pc/l for concentrations between 50 and 100 pc/l.

e. Uses

This procedure is of particular value when doing routine, day to day, analysis of samples with a consistent radionuclide content.

D. Least Squares Analysis

1. GENERAL

Although not in major use at present on a routine basis, this method provides a procedure for the accurate analysis of complex gamma spectra at environmental levels. It is a method specifically designed for computer application.

2. EVALUATION

a. Advantages

1. The activity of each radionuclide is calculated using the data from each channel rather than a selected photopeak area. Also, the counting error can be calculated for each nuclide.

2. There are no subjective errors involved.

3. An analysis can be made for peaks which are superimposed.

4. A correction for nuclides not accounted for in the original library of standards can be made.

b. Disadvantages

1. It cannot be executed without a computer capability.

2. It requires a stable instrument for realistic results.

c. Precision

For low activity samples (environmental levels) accuracy and precision within .5-2% can be expected.

IV. SUMMARY

A summary of the precision and minimum detectability of the different methods available for quantitative gamma analysis are given in Table 1.

Table 1. Comparison of analyses methods

Method	Minimum detectability at 50 min. count	% Error at 100 pc/l	Uses
Radiochemical Separation plus Analysis	10 pc/l	± 10%	Single Channel Analysis
Compton Continuum Subtraction	10-30 pc/l	± 10-30%	Routine Estimated Concentrations
Spectrum Stripping	10 pc/l	± 10%	Spot check - Routine with Spectrum Stripper
Simultaneous Equations	10 pc/l	± 10%	Routine analysis Computer Application
Least Squares Analyses	---------	± .5-2%	Computer Application

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GPO 886-421

Sequential Assembly of Gamma Spectrometer Systems

I. INTRODUCTION

In assembling an instrument system for gamma analysis, both the immediate and long range objectives of the program must be defined. The immediate objective will be to provide a system capable of satisfying present analytical commitments, (i.e., number of samples to be analyzed, minimum sensitivity desired, and degree of accuracy necessary), as far as is economically possible. The long range objectives will take into consideration expansion of responsibility or intensification of the program resulting from increased levels of activity. Some of the factors affecting the sequence of purchasing components, and a cost analysis of representative systems are considered.

II. SEQUENCE OF PURCHASE

Although it would be ideal to provide the capability for meeting both the immediate and long range objectives in one purchase, it is usually not economically feasible. The selection of equipment for the initial purchase then should meet two requirements. First, it must provide a system whicn meets both qualitatively and quanitatively, the immediate objectives of the program and second it must be capable of expansion.

Where the initial gamma analysis load does not justify a multichannel analyzer, the sodium iodide crystal and shield should receive priority as they are more difficult to up-grade than the associated counting instrumentation. A 3" × 3" or 4" × 4" crystal and a 6" thick steel shield provide a good system for even the "sophisticated" analysis techniques.

An inexpensive single-channel system can be arranged by using an IPC or GM scaler and high voltage supply with a pulse height analyzer module. Although the crystal and shield are not being used to their full capability, it is much more desirable than having a multichannel analyzer connected to a small crystal and shield inadequate for gamma spectral analysis of environmental samples. The multichannel analyzer can be substituted for the single channel analyzer as the need and funds dictate.

The purchase of read-out equipment and its compatibility with computer programming and information input techniques is also important in terms of long range expansion.

III. COST ANALYSIS

The following is a cost analysis indicating the approximate cost of various components of gamma spectrometer systems and of several typical complete systems. The prices shown are indicative of the 1964 commercial list prices of manufacturers in the nuclear instrumentation field. Obviously, the cost of a specific make or model may vary significantly from that listed due to the design, quality, and total function of the component.

A. Scintillation Detector - (Tl) crystal, photomultiplier tube, and pre-amplifier.

1. Crystal - price is dependent on size and configuration of crystal.

COST ANALYSIS OF NaI(Tl) CRYSTALS

Crystal Size	Cost
½" × ½"	$30.00
3/4" × 3/4"	35.00
1" × 1"	50.00
1½" × 1"	95.00
1½" × 1½"	130.00
1½" × 2"	170.00
2" × 1"	160.00
2" × 2"	270.00
3" × 2"	550.00
3" × 3"	750.00
4" × 2"	860.00
4" × 4"*	1500.00
5" × 5"*	2300.00
7" × 5"*	4300.00
9" × 4"*	5500.00
1-3/4" × 2" (well)	250.00
2" × 2" (well)	320.00
3" × 3" (well)	850.00

*Matched Window Type

2. Photomultiplier tube - may be bought as a separate unit or in combination with the crystal. Cost is dependent on the size, sensitivity, and gain characteristics of the tube.

COST ANALYSIS OF PHOTOMULTIPLIER TUBES

For Crystal Size	Avg. Sensitivity	Approx. Gain	Approx. Cost
1"	60 μa/lumen	10^6	$50 - $ 80
2"	"	"	50 - $ 80
3"	"	"	100 - $150
5"	"	"	150

3. Pre-amplifiers - bought as a separate unit.

Cost $100 - $200

4. Additional equipment

a. External high flux shield $20 - $35.

b. Low background base assembly $75.

c. Copper or stainless steel crystal container $100.

B. Shielding

1. For single channel analysis utilizing crystals up to 3" × 3".

Equivalent shield - 2" Pb.

Cost $600

2. For single channel analysis utilizing crystals up to 5" × 5".

Equivalent shielding - 2.5" - 3".

Cost $1000

3. For low level multi-channel analysis involving environmental samples 6" steel in all directions.

Dimensions	Cost
20" × 20" × 24"	$4000
24" × 24" × 36"	5000

C. Single Channel Analyzers

COST ANALYSIS FOR COMPONENTS OF SINGLE CHANNEL ANALYZERS

Item	Cost
Linear Amplifier	$ 500 - 700
Pulse Height Analyzer	$ 500 - 700
Scaler	$ 500 - 700
High Voltage Supply	$ 400 - 600
Timer	$ 100 - 500
Total Basic Unit Less detector and shielding	$2000 - 3200

Extras

Ratemeter	$ 500 - 800
Spectrum Scanner	$ 500 - 700
Total	$1000 - 1500

For entire single channel system including probe detector and shielding. (See Figure 1)

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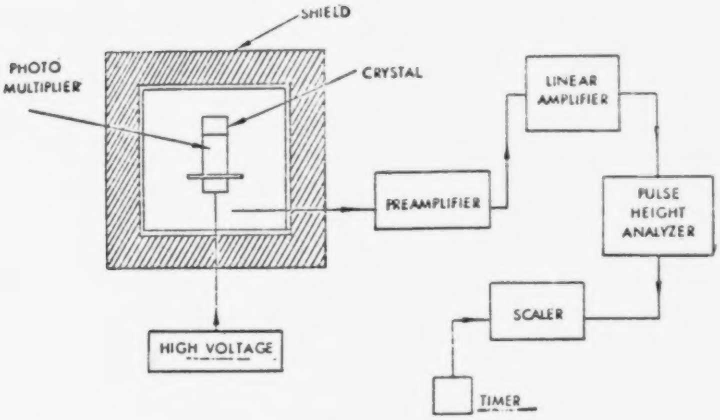

ESSENTIAL SINGLE CHANNEL ANALYZER COMPONENTS

Figure 1

Item	Cost
Total Base Unit, less detector and shielding	$2000 - 3200
Shielding	$4000
Probe Detector	$1000
Extras	$1000 - 1500
Total	$7000 - 10,000

D. Multichannel Analyzers

Most instruments include the following:

1. Linear amplifier
2. High voltage supply
3. Pulse height analyzer
4. Data memory
5. Timer and scaler
6. Cathode display
7. Pulse Generator

COST ANALYSIS OF MULTICHANNEL ANALYZERS

Analyzer	Cost
100 channel	$ 6,000
400 channel	$ 9,000 - 12,000
800 channel	$15,000 - 18,000
1000 channel	$20,000
4000 channel	$30,000

Price range for given number of channels depends on the number of inputs and storage capacity.

Each additional input	$400
Additional storage, 10^x	$700 per x
Differential accumulation	$750

Prices are for transistor analyzers with analog to digital conversion.

E. Readout Indicators

The cost of a readout display system depends on the type of display desired and the readout from the analyzer. The readout is usually in digital or analog form, enabling direct readout to an X-Y plotter, tape printer, or oscilloscope. A converter must be used however, to transform the digital output in parallel form to serial form in order to utilize a typewriter or tape punch unit. (See Figure 2)

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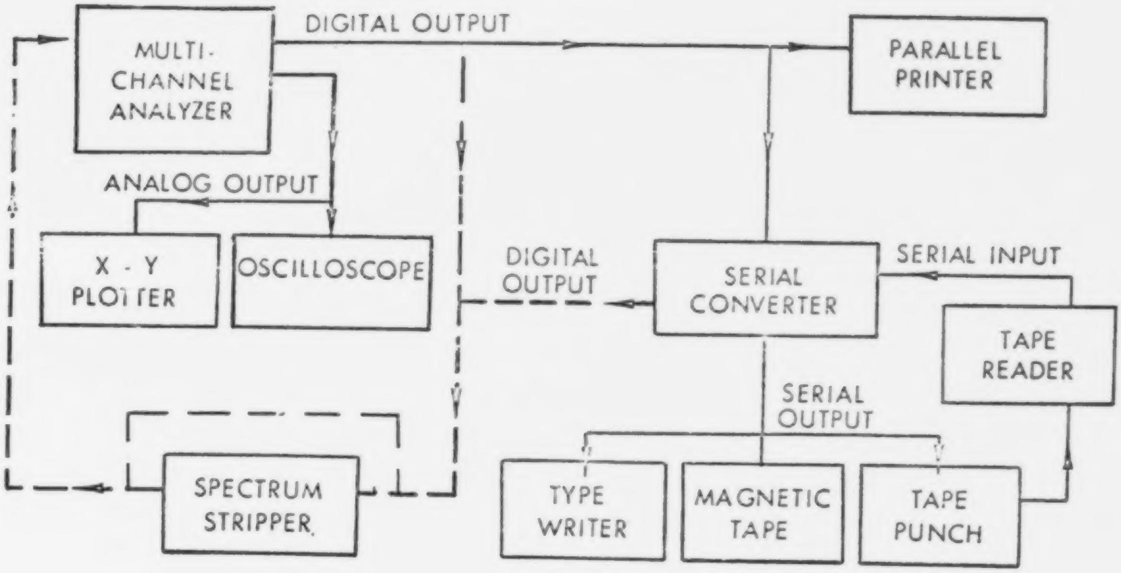

MULTI-CHANNEL ANALYZER
DIGITAL OUTPUT
PARALLEL PRINTER
ANALOG OUTPUT
X - Y PLOTTER
OSCILLOSCOPE
DIGITAL OUTPUT
SERIAL CONVERTER
SERIAL INPUT
TAPE READER
SERIAL OUTPUT
SPECTRUM STRIPPER
TYPE WRITER
MAGNETIC TAPE
TAPE PUNCH

TYPICAL READOUT SYSTEM

Figure 2

COST ANALYSIS OF READOUT INDICATORS

Item	Cost
Oscilloscope	$ 700
Oscilloscope Camera	$ 400
Tape Printer	$1500 - $2000
X - Y Plotter	$2500 - $2700
Serial Converter	$1200 - $4000
IBM Readout Typewriter	$1000
Tape Puncher	$1200
Tape Reader	$1000
Tape Control Unit	$1000
Magnetic Tape Unit	$1200
Extras	
Tape to Typewriter Converter	$1000
Spectrum Stripper	$2500

400 CHANNEL SYSTEM

Item	Cost
400 Channel Analyzer	\$ 9,000 - \$12,000
Shielding	\$ 4,000 - \$ 5,000
Probe	\$ 1,000
Readout	
Tape Printer	\$ 1,500 - \$ 2,000
Total	\$15,000 - \$20,000
Additional for Spectrum Stripping	
Serial Converter	\$ 1,200 - \$ 4,000
Tape Puncher and Reader	\$ 2,200
Spectrum Stripper	\$ 2,500
Total	\$ 5,900 - \$ 8,700

IV. OTHER CONSIDERATIONS

A. Service of Instruments

B. Warranty

C. Trial Periods for Testing Instrument

D. Ordering Procedure - always specify in writing, the specifications for the instruments being ordered.

V. SUMMARY

In selecting an instrument system for gamma analysis, two factors must be considered. First, it is necessary to establish both the quantitative and qualitative criteria which the system must satisfy. Second, a cost analysis of the equipment necessary to meet the established criteria must be made. From this it is possible to define to what extent the program may be fulfilled in view of financial capabilities. Finally, a logical building block program may be initiated to provide for easy expansion of the initial system.

Characteristics of Gamma Spectra Problem Session

I. INTRODUCTION

The purpose of this problem session is to introduce the trainee to several characteristics of gamma spectra which are applied to various methods of qualitative and quantitative spectral analysis.

II. PROCEDURE

A. Standard Spectra

Figures I, II, and III are gamma spectra of single radionuclides. Determine what nuclide each spectrum represents by comparison to standard spectra in the "Gamma Spectrum Catalogue". Report results in Table 1.

Table 1

Spectra	Nuclide
Figure I	
Figure II	
Figure III	

B. Spectral Shape

Figures IV, V, and VI are gamma spectra for scandium-46, manganese-54, and sulfur-37 respectively. Determine the cause (gamma photons, x-rays, Compton peaks, etc) for each peak in the spectra. Decay schemes are included. Report data in Table 2.

Table 2

Scandium-46		Manganese-54		Sulfur-37	
Energy of peak	Cause	Energy of peak	Cause	Energy of peak	Cause

C. Constant Spectra Shape

A sample of ruthenium-103 containing 355 pc was counted and produced the gamma spectrum plotted in Figure VII (digital data given in Table 3). Determine the spectrum which would result if a sample containing 890 pc of Ru^{103} was counted on the same system. Plot the spectrum on Figure VII.

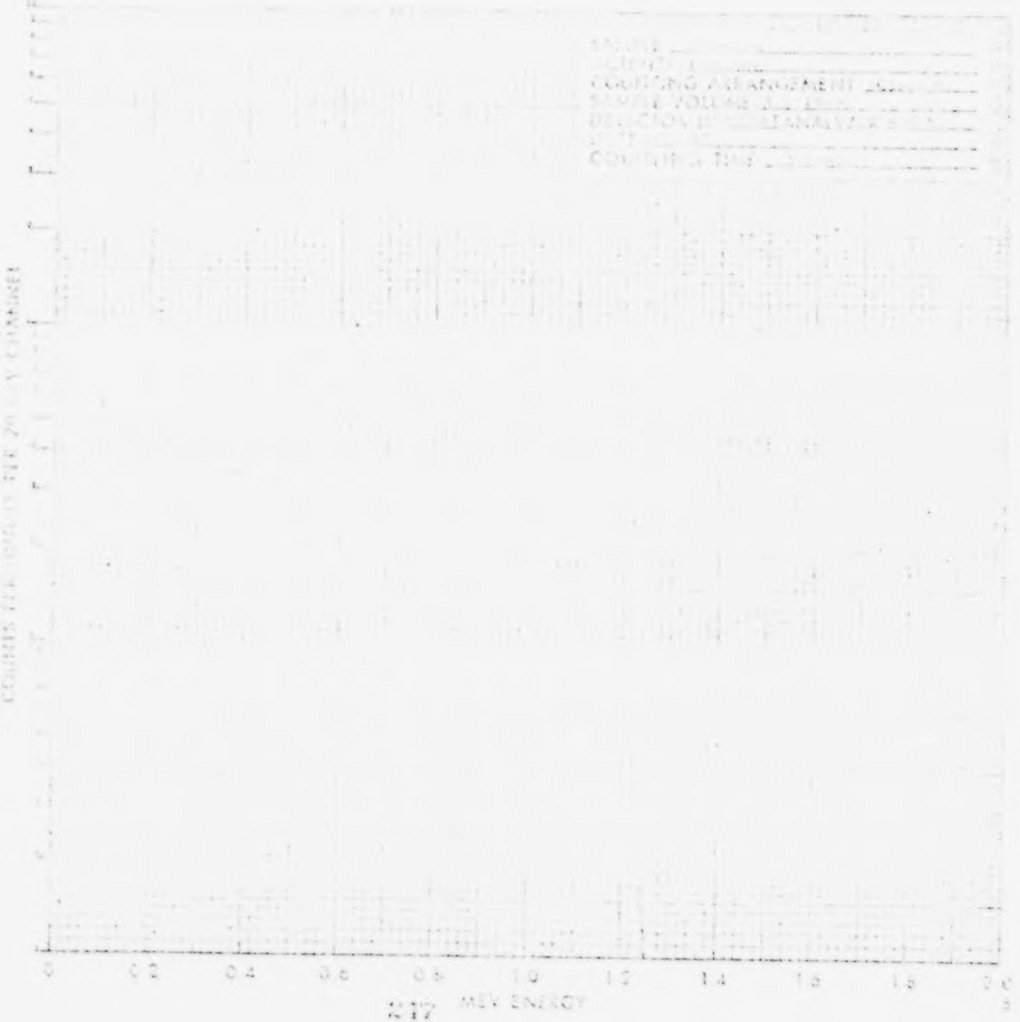
Table 3

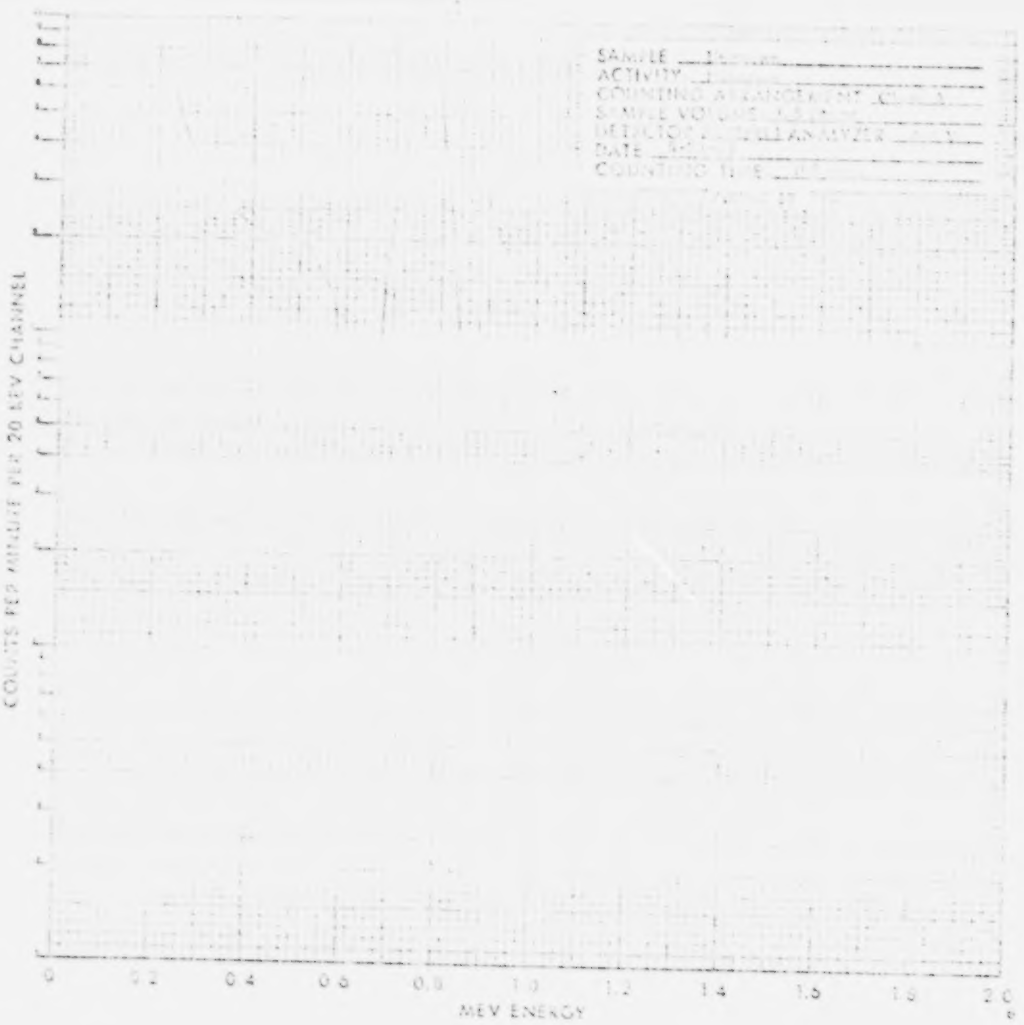
Energy	Count rate (355 pc)	Count rate (890 pc)
.08	23.1	
.12	25.8	
.16	27.2	
.20	36.0	
.24	32.3	
.28	30.5	
.32	20.1	
.36	13.2	
.40	12.1	
.44	20.9	
.48	160.0	
.52	150.3	
.56	12.7	
.60	19.9	
.64	6.1	
.68	1.1	

D. Standard spectra of Cs^{137} (500 pc) and I^{131} (500 pc) are plotted in Figure VIII (digital data given in Table 4). Determine the spectrum of a sample, counted on the same system, which contains a mixture of 500 pc of Cs^{137} and 500 pc of I^{131}. Plot the spectrum on Figure VIII.

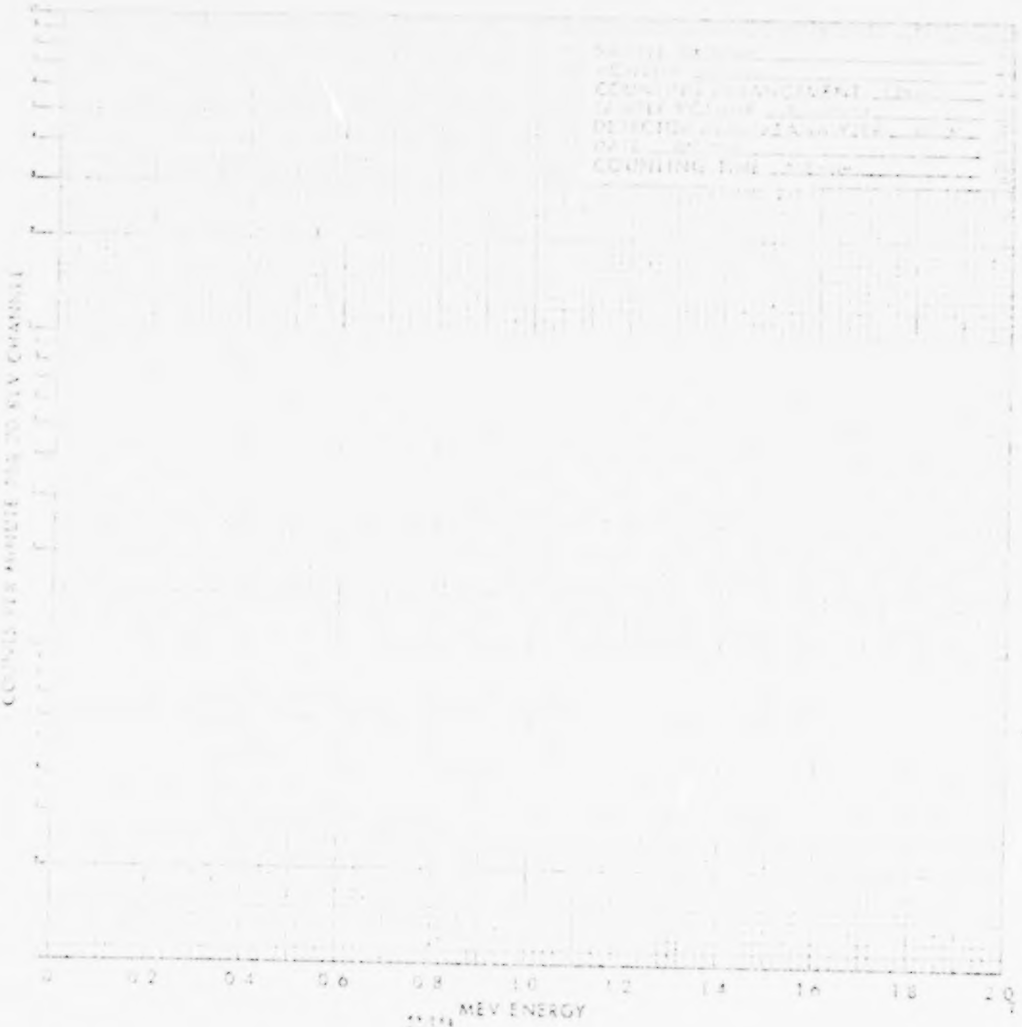
Table 4

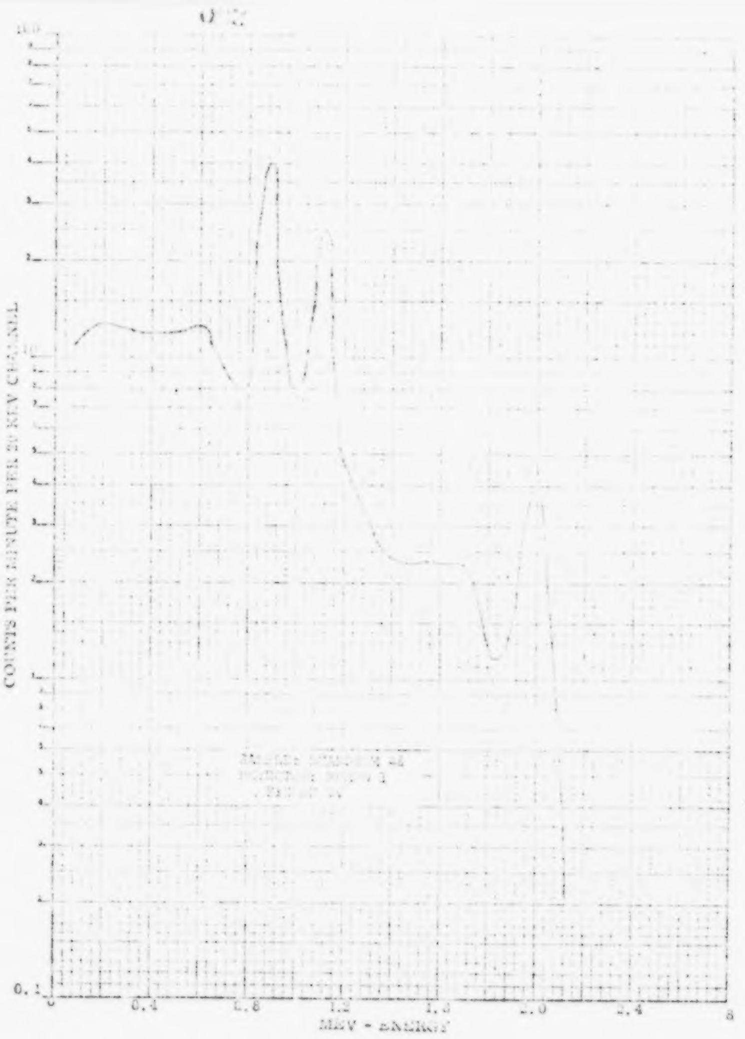
Energy	Count rate 500 pc I^{131}	Count rate 500 pc Cs^{137}	Count rate 500 pc I^{131} + 500 pc Cs^{137}
0.1	82.1	301.2	
0.15	96.3	300.1	
0.20	46.0	349.9	
0.25	38.1	2[illegible].0	
0.30	36.2	219.9	
0.35	160.0	190.0	
0.40	32.1	180.2	
0.45	4.3	150.1	
0.50	5.0	110.9	
0.55	2.8	50.1	
0.60	5.4	170.1	
0.65	4.2	580.2	
0.70	4.2	220.1	


SAMPLE
ACTIVITY
COUNTING ARRANGEMENT
SAMPLE VOLUME
DETECTOR
ANALYZER
COUNTING TIME
COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY
N-17



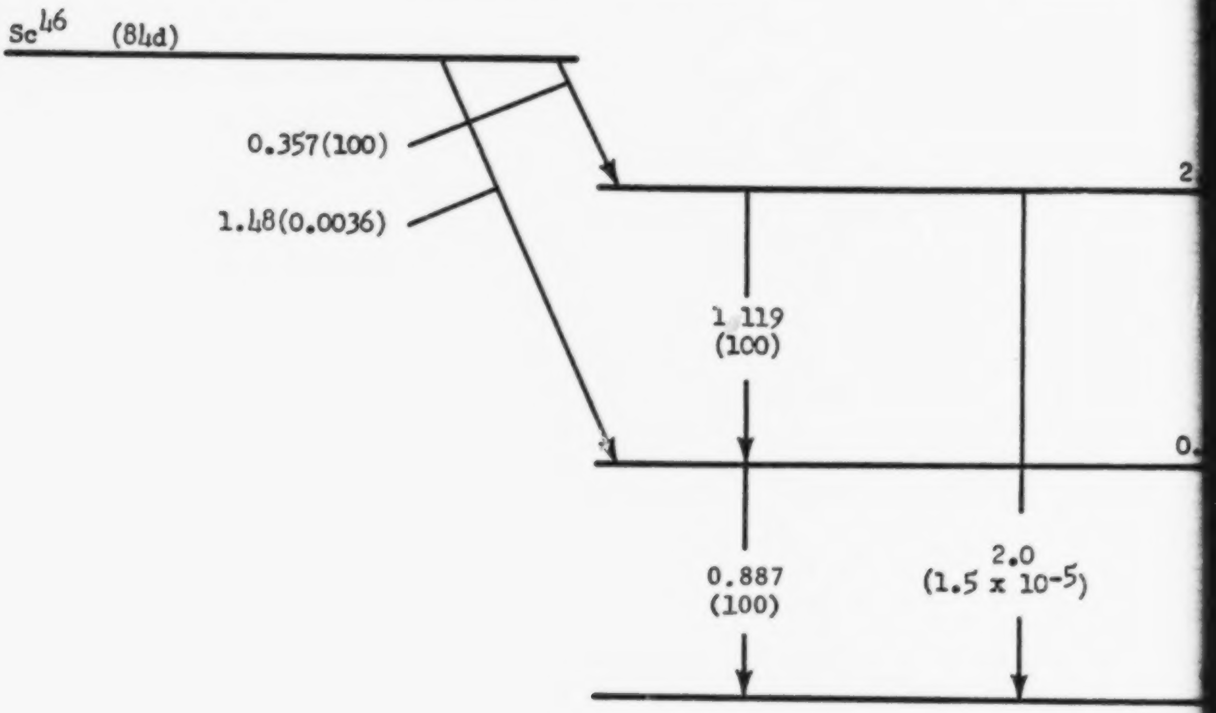
SAMPLE [illegible]
ACTIVITY [illegible]
COUNTING ARRANGEMENT [illegible]
SAMPLE VOLUME [illegible]
DETECTOR [illegible] ANALYZER [illegible]
DATE [illegible]
COUNTING TIME [illegible]


COUNTING ARRANGEMENT
SAMPLE VOLUME
DETECTOR
ANALYZER
DATE
COUNTING TIME
COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY

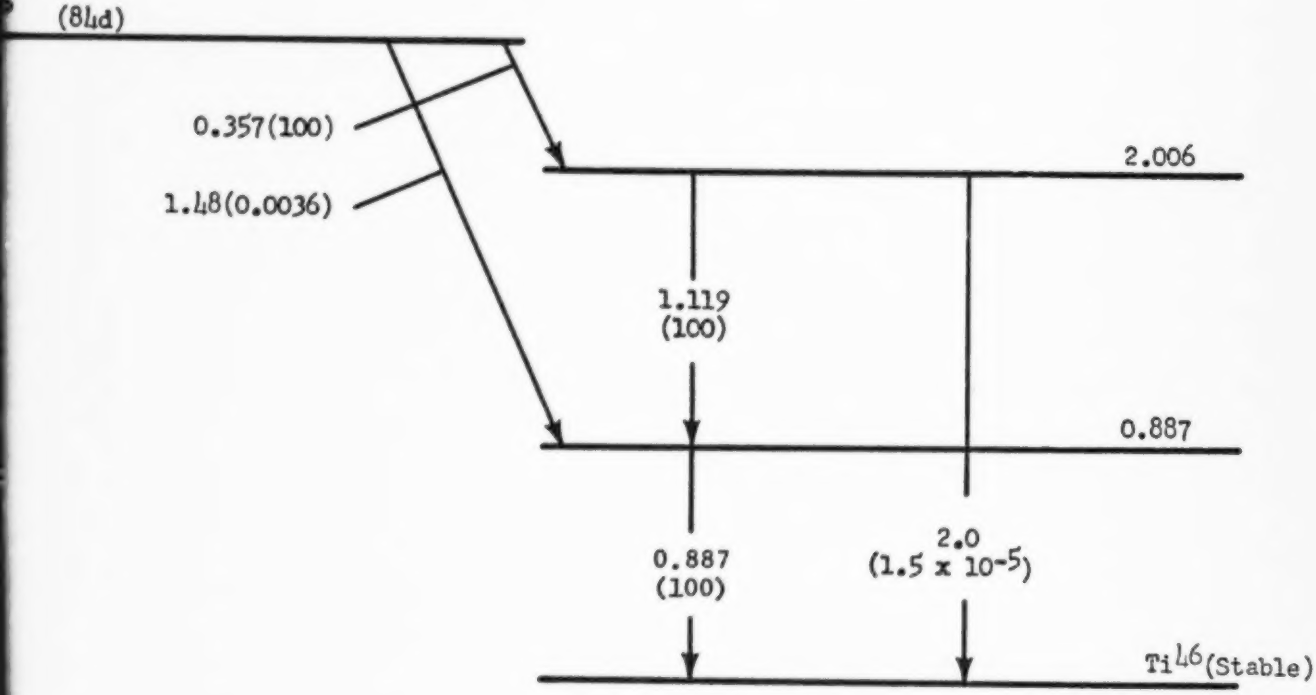

COUNTS PER MINUTE PER 20 KEV CHANNEL
100
10
1
0.1
0
0.4
0.8
1.2
1.6
2.0
2.4
8
MEV - ENERGY

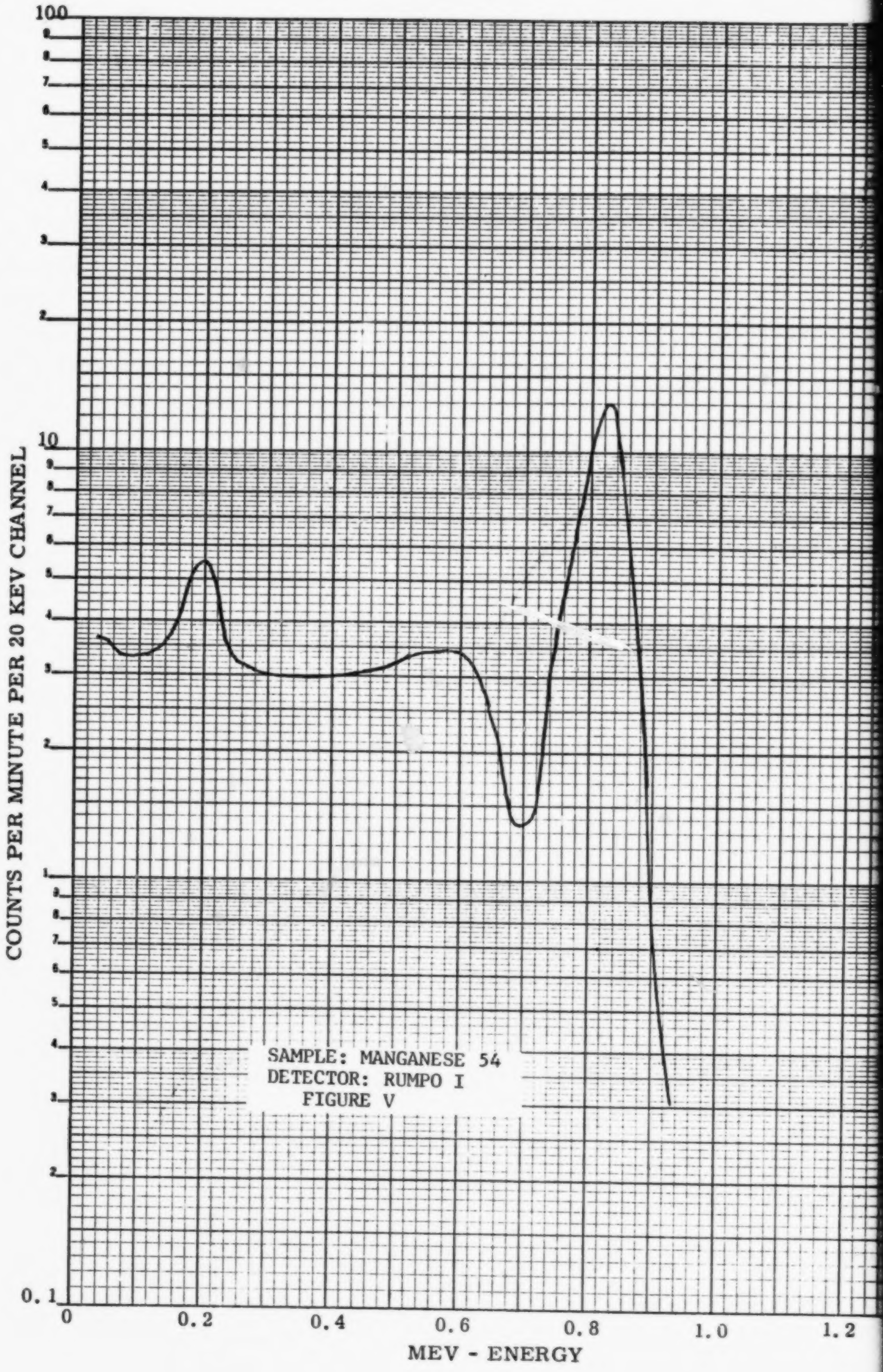
Scandium-46 DECAY SCHEME

December 23

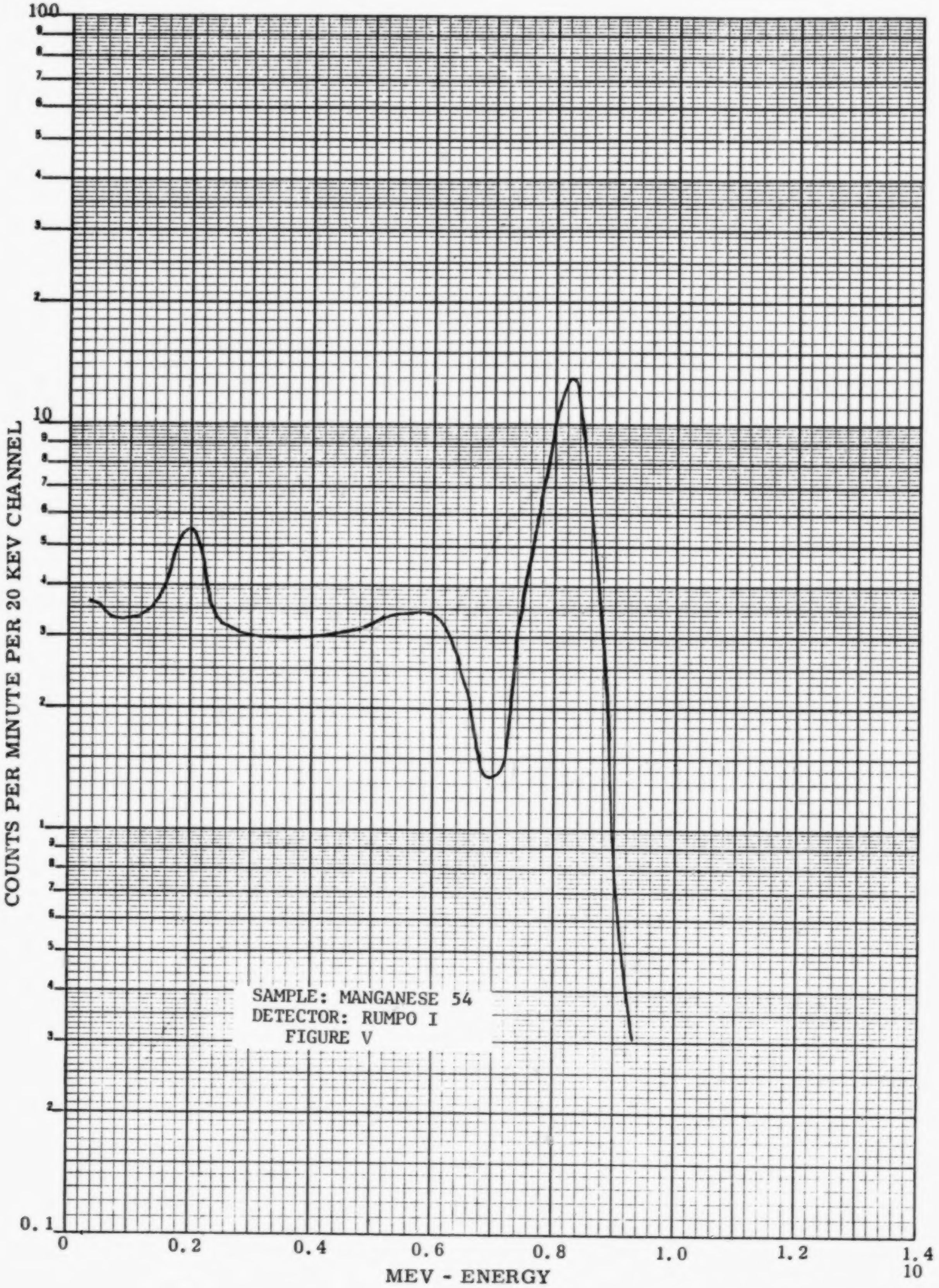

Sc^{46} (84d)
0.357(100)
1.48(0.0036)
2
1.119
(100)
0.
0.887
(100)
2.0
(1.5 x 10^{-5})

Scandium-46 DECAY SCHEME December 23, 1963

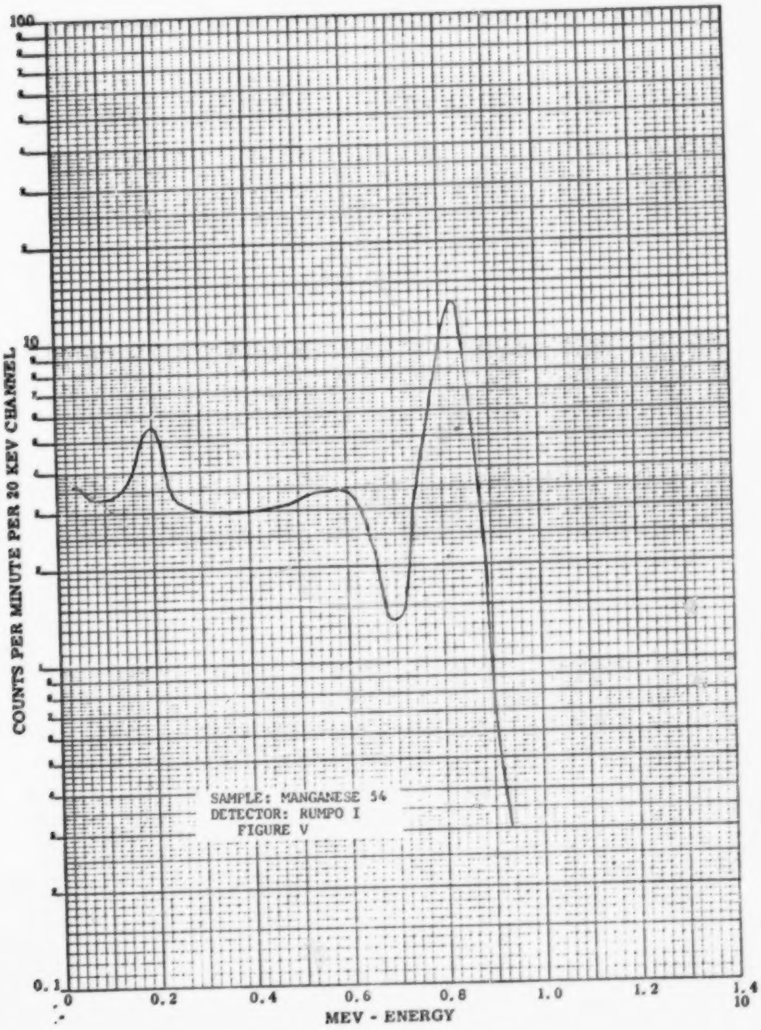

(84d)
0.357(100)
1.48(0.0036)
2.006
1.119
(100)
0.887
0.887
(100)
2.0
(1.5 x 10-5)
Ti46(Stable)



SAMPLE: MANGANESE 54
DETECTOR: RUMPO I
FIGURE V

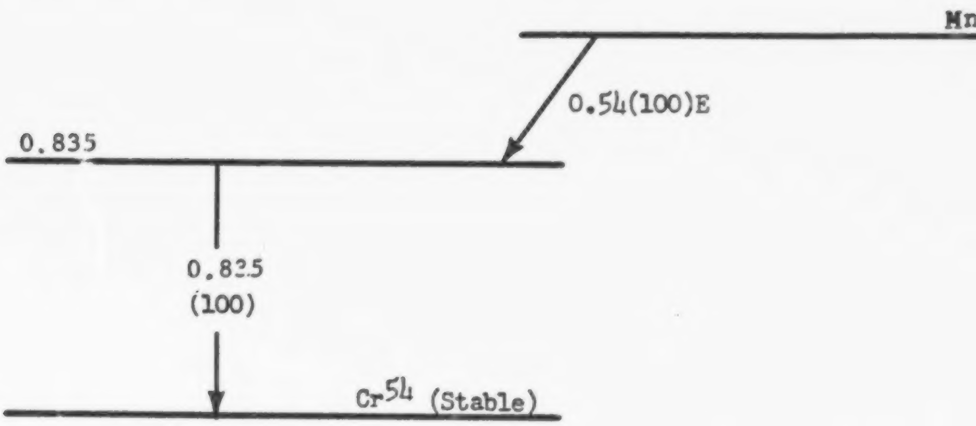


SAMPLE: MANGANESE 54
DETECTOR: RUMPO I
FIGURE V



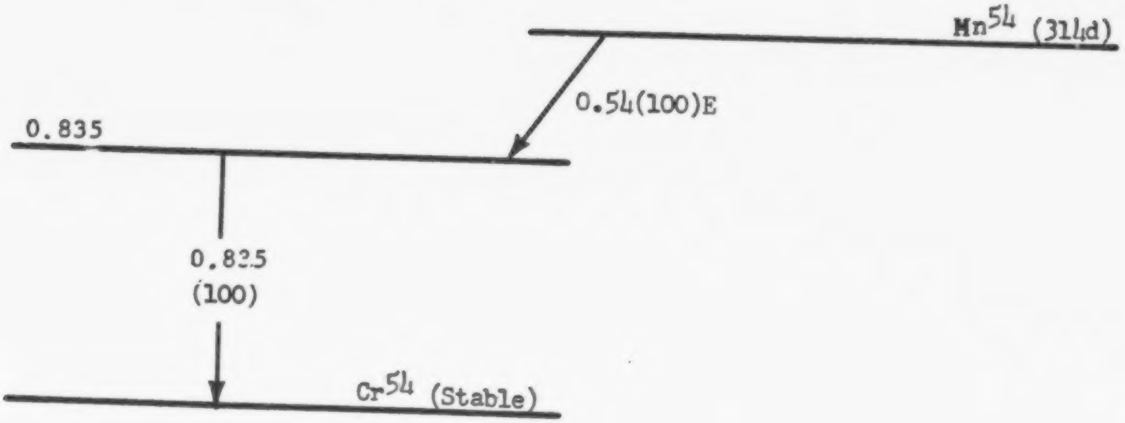
SAMPLE: MANGANESE 54
DETECTOR: RUMPO I
FIGURE V

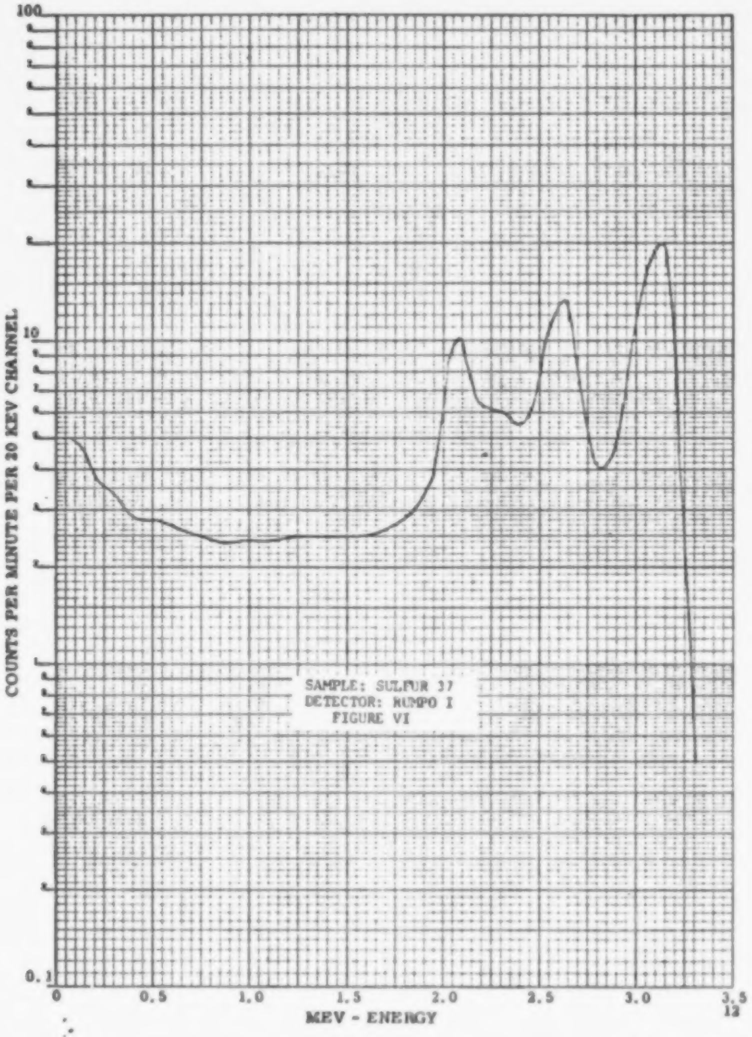
Manganese-54 DECAY SCHEME


Mn
0.54(100)E
0.835
0.825
(100)
Cr54 (Stable)

A

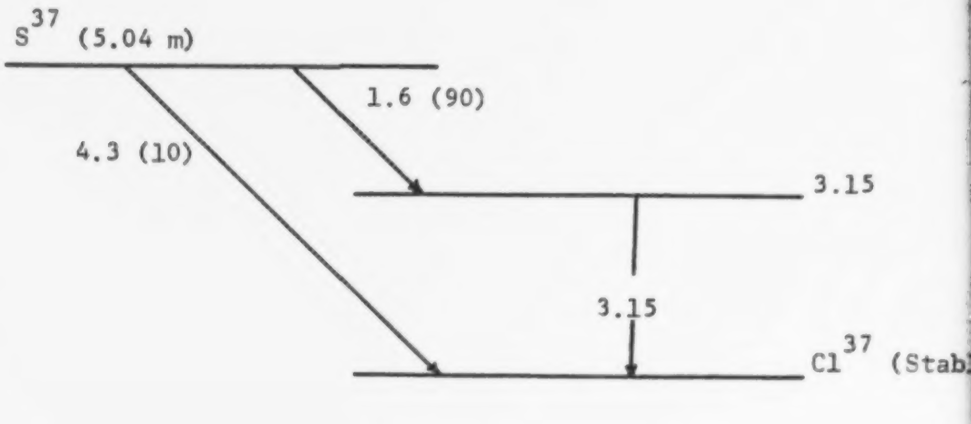
Manganese-54 DECAY SCHEME 12/12/63


Mn^{54} (314d)
0.54(100)E
0.835
0.835
(100)
Cr^{54} (Stable)



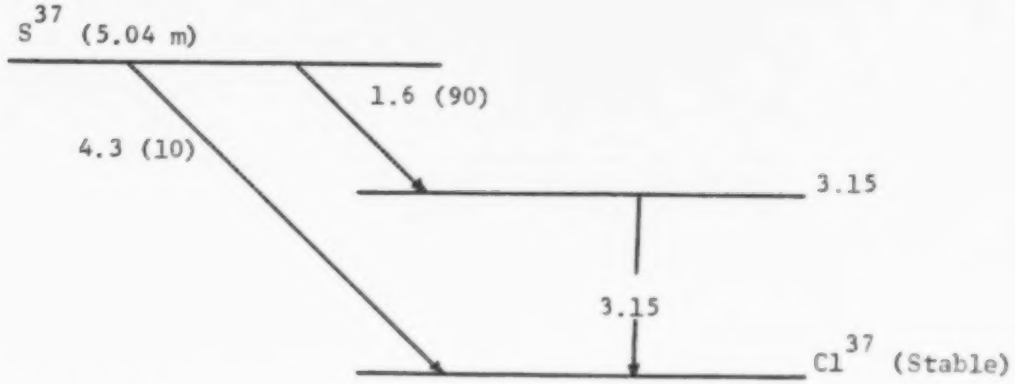
SAMPLE: SULFUR 37
DETECTOR: HUMPO I
FIGURE VI

SULFUR - 37 DECAY SCHEME


S^{37} (5.04 m)
1.6 (90)
4.3 (10)
3.15
3.15
Cl^{37} (Stab

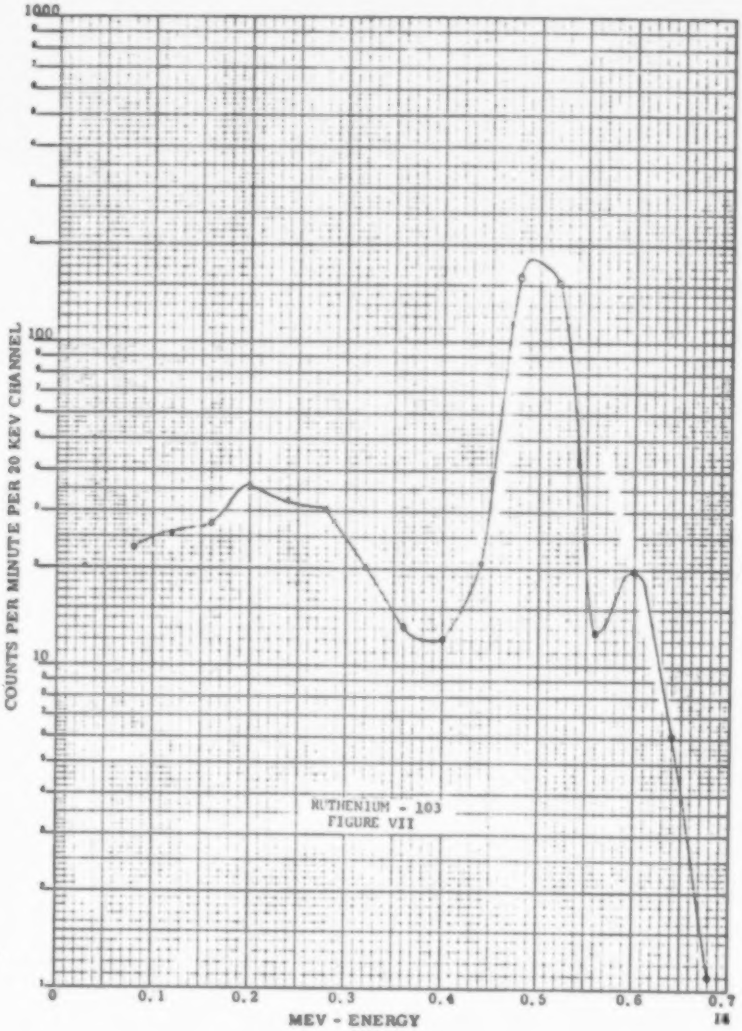
A

SULFUR - 37 DECAY SCHEME



S^{37} (5.04 m)
1.6 (90)
4.3 (10)
3.15
3.15
Cl^{37} (Stable)

B



RUTHENIUM - 103
FIGURE VII

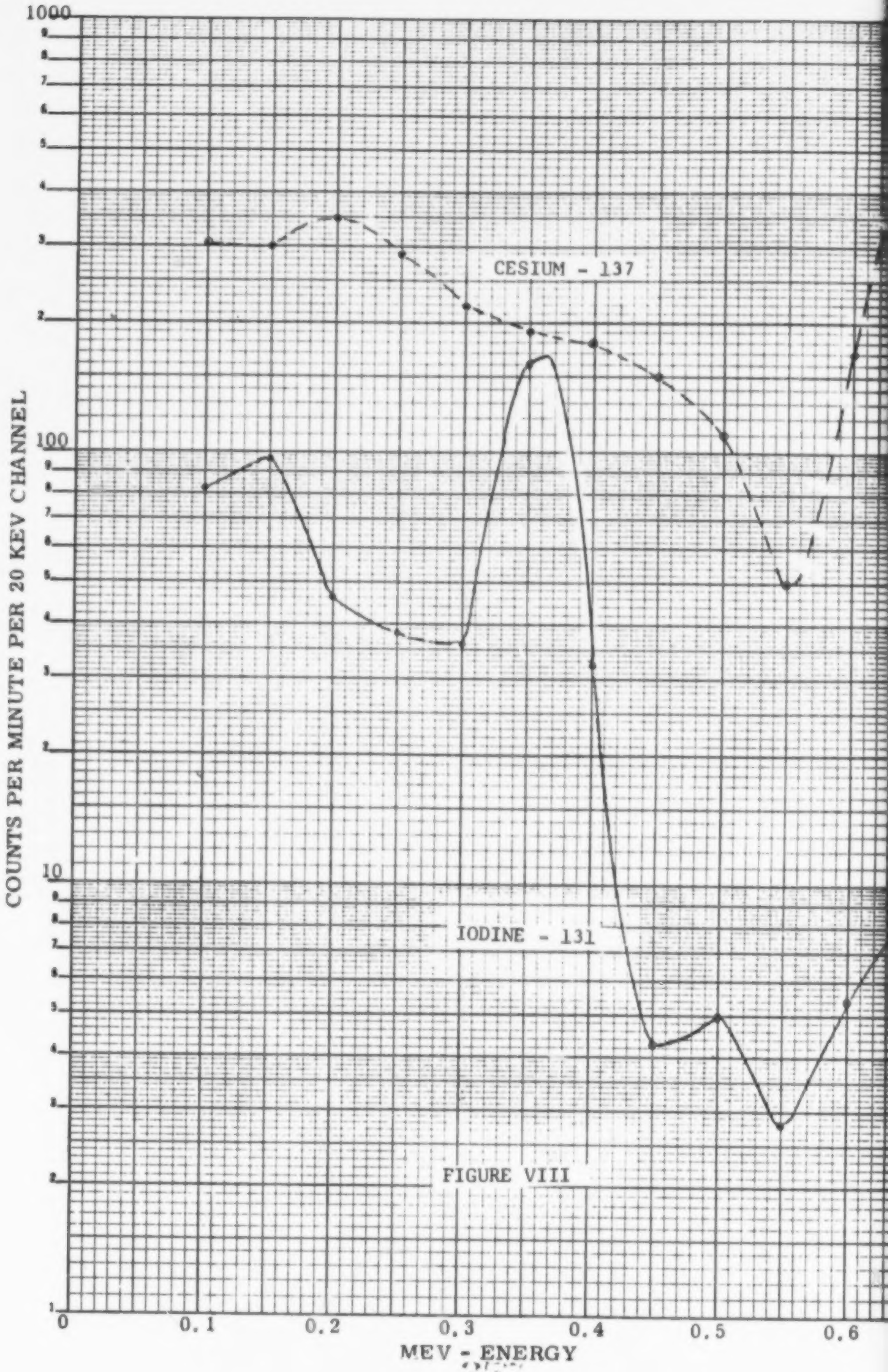


FIGURE VIII

257

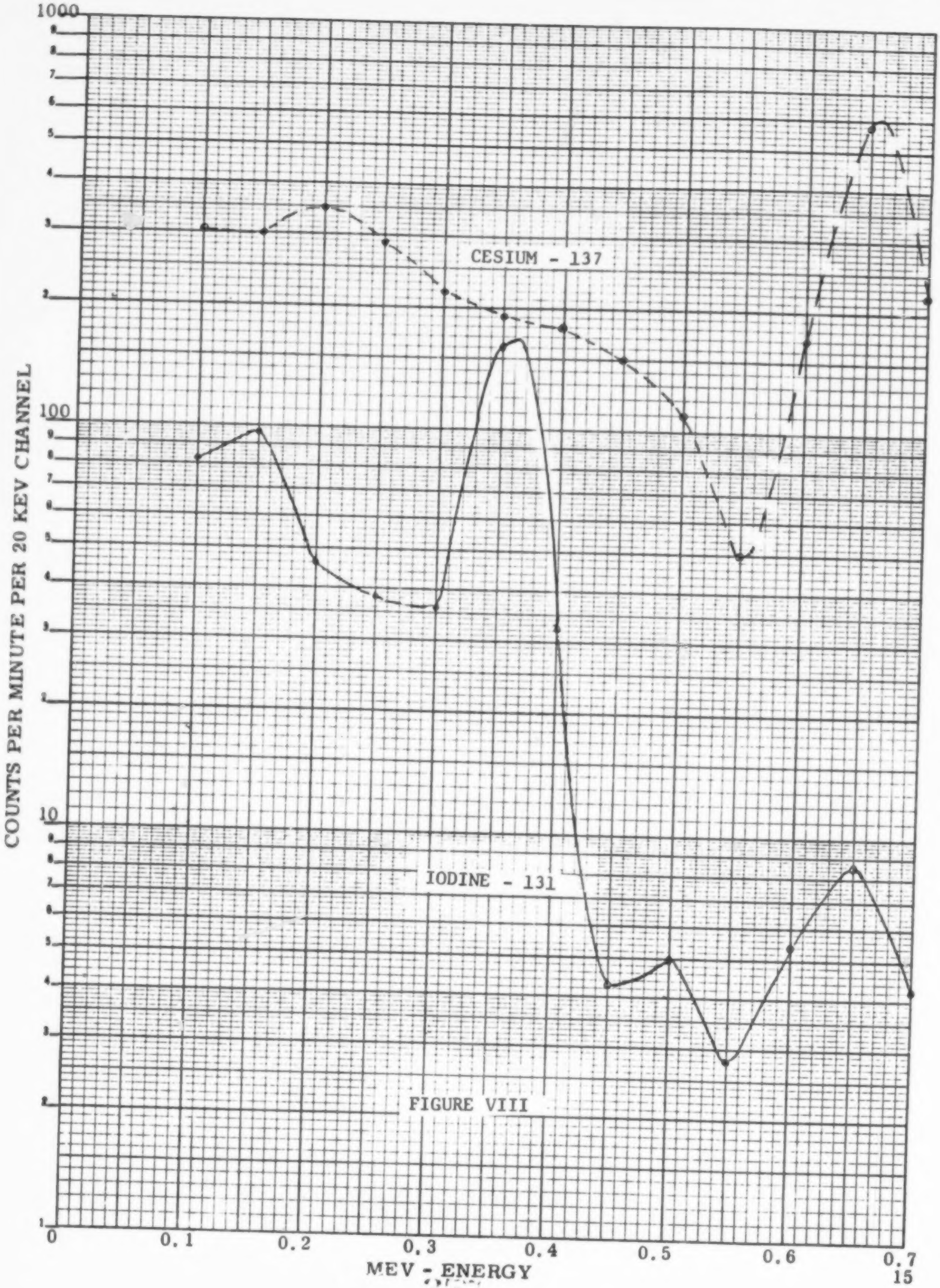

1000
100
10
1
CESIUM - 137
IODINE - 131
COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
MEV - ENERGY

FIGURE VIII

Energy Calibration
Problem Session

I. INTRODUCTION

This problem session is designed to give the trainee experience in the calculation of channel coefficient as utilized in the energy calibration of a gamma spectrometer. Both the graphical and mathematical procedure will be utilized in determining fractional peak channels.

II. STATEMENT OF PROBLEM

Two calibration spectra have been obtained. The operator, however, failed to record the energy calibration he was using on each spectrum. Before any analysis can be performed, it is necessary to determine what channel coefficients were used.

III. PROCEDURE

A. Mathematical Procedure

Determine the channel coefficients for spectra given in Figures 1 and 2. Use the accompanying data to determine the actual fractional peak channel to within 0.1 of a channel.

B. Graphical Procedure

Determine the channel coefficient for spectrum given in Figure 1. Use the accompanying data to determine the actual fractional peak channel to within 0.1 of a channel.

IV. RESULTS

CHANNEL COEFFICIENT

A. Mathematical Procedure

Nuclide	Photopeak Energy	Photopeak Channel	Channel Coefficient
Cs^{137} - Fig. I			
Cs^{137} - Fig. II			

B. Graphical Procedure

Nuclide	Photopeak Energy	Photopeak Channel	Channel Coefficient
Cs^{137} - Fig. I			

SAMPLE [illegible]
ACTIVITY [illegible]
COUNTING ARRANGEMENT [illegible] A
SAMPLE VOLUME 3.5 liter
DETECTOR [illegible] ANALYZER [illegible]
DATE [illegible]
COUNTING TIME: 100 min.

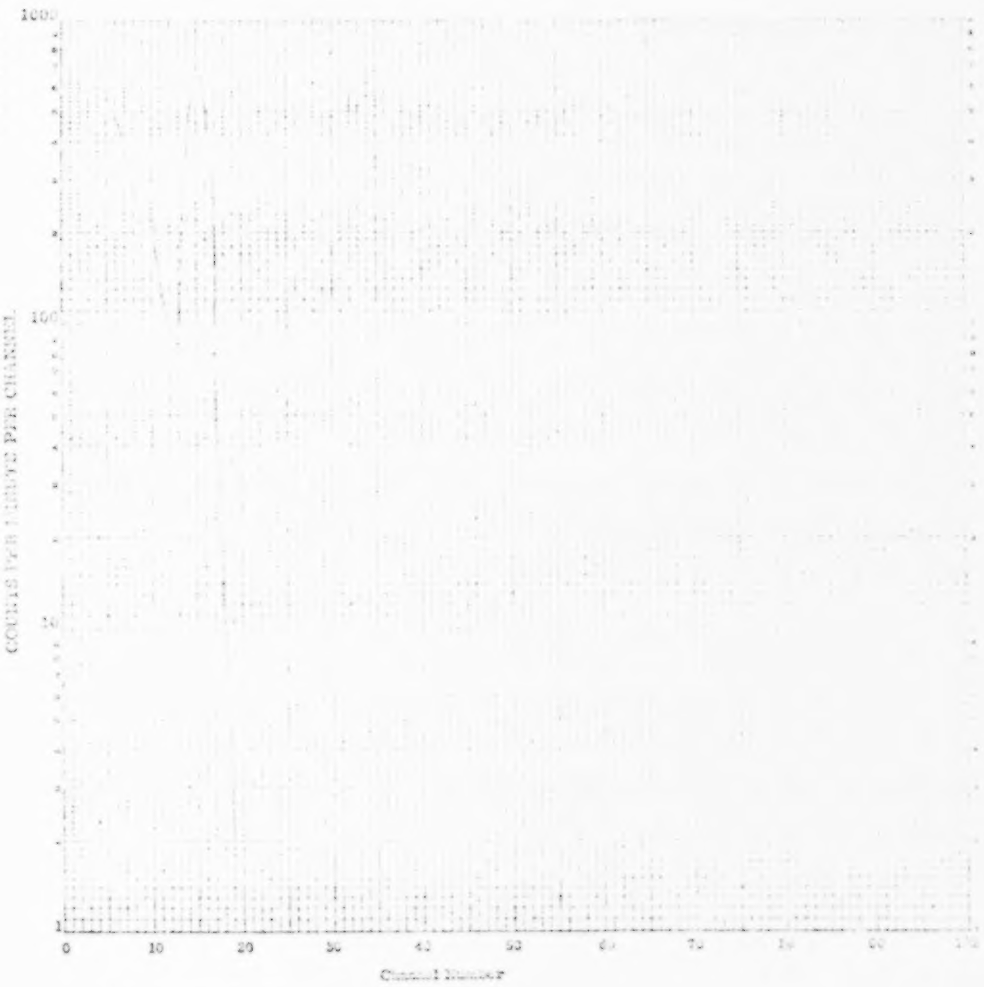


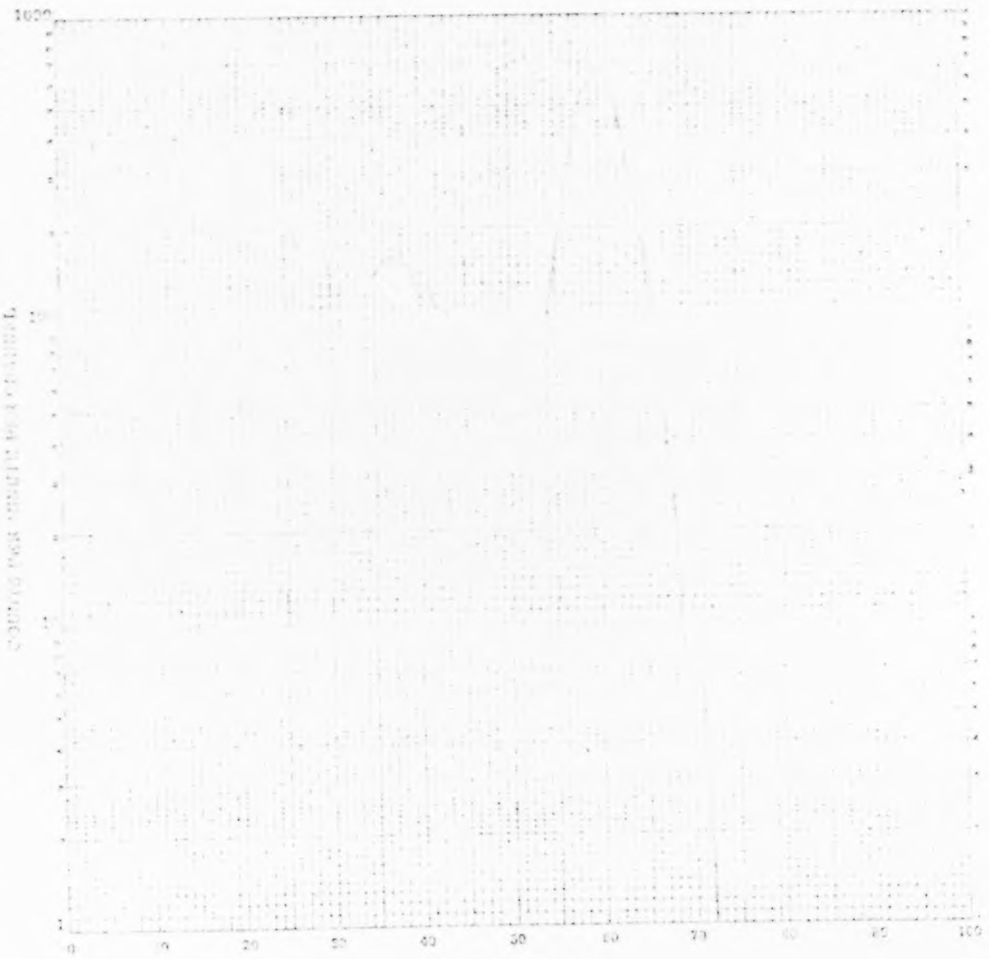
Figure 1

SAMPLE: Cs-137

ACTIVITY: [illegible]

COUNTING ARRANGEMENT: [illegible]

COUNTING TIME: [illegible]



Channel Number

FIGURE 2

22150

Bureau of Radiological Health

Evaluation of Linearity and Stability

Problem Session

I. INTRODUCTION

This problem session is designed to give the trainee experience in the procedures for evaluating the linearity and stability of a spectrometer system.

II. STATEMENT OF PROBLEM

A. Linearity

In part A, it is desired to determine the integral linearity of the total spectrometer system. The instrument has been calibrated to 0.2 Mev/channel using a 0.145 Mev Ce^{141} photopeak and a 1.85 Mev Y^{88} photopeak. If the instrument displays a linear response, a graph of photopeak channel versus gamma energy for a series of nuclides of various energies will yield a straight line between the calibration points. Develop such a graph and observe any deviation from the theoretical linearity.

B. Stability

In part B, data have been gathered for two weeks which represents the Cs^{137} photopeak with a constant gain setting. From this data it is required to check the long term stability of the instrument, i.e., to determine if the Cs^{137} photopeak falls in the same channel and if not how much drift is observed.

III. PROCEDURE

A. Determine the linearity of the total system.

1. Determine the fractional photopeak channel of Y^{88} and Ce^{144} using the data given in Table 1 and check the calibration. Draw theoretical response curve.

2. Graph actual photopeak channel versus true energy from data given in Table 1.

3. Graph channel deviation versus channel number for same nuclides.

4. Determine the maximum channel deviation from the theoretical value.

5. Determine integral linearity using the maximum channel deviation and a maximum rated energy equal to 2 Mev (100 channels at .02 Mev/ch).

B. Determination of Instrument Drift

1. Plot actual peak channel given in Table 2 versus time.

2. Draw line of best fit.

3. Compute instrument drift in channels per week.

Table 1

DATA FOR PART A

Y^{88} - 1.85 Mev

Channel No.	Net cpm
91	7.75
92	8.54
93	8.57
94	7.78

Ce^{141} - 0.145 Mev

Channel No.	Net cpm
06	719.82
07	935.31
08	1216.30
09	280.79

R/N	E	Actual Peak Channel	Peak Channel at .02 Mev/ch	Channel Deviation
Y^{88}	1.85			
K^{40}	1.46	72.7		
Zn^{65}	1.14	56.5		
Y^{88}	.889	44.6		
Cs^{137}	.662	32.9		
Cr^{51}	.320	15.9		
Ce^{141}	.145			

IV. RESULTS

A. Linearity

1. Maximum channel deviation from theoretical = ______________ channels

2. Integral linearity = ______________

B. Stability

1. Instrument drift = ______________ channels/week

Table 2

DATA FOR PART B

Nuclide: Cs^{137}

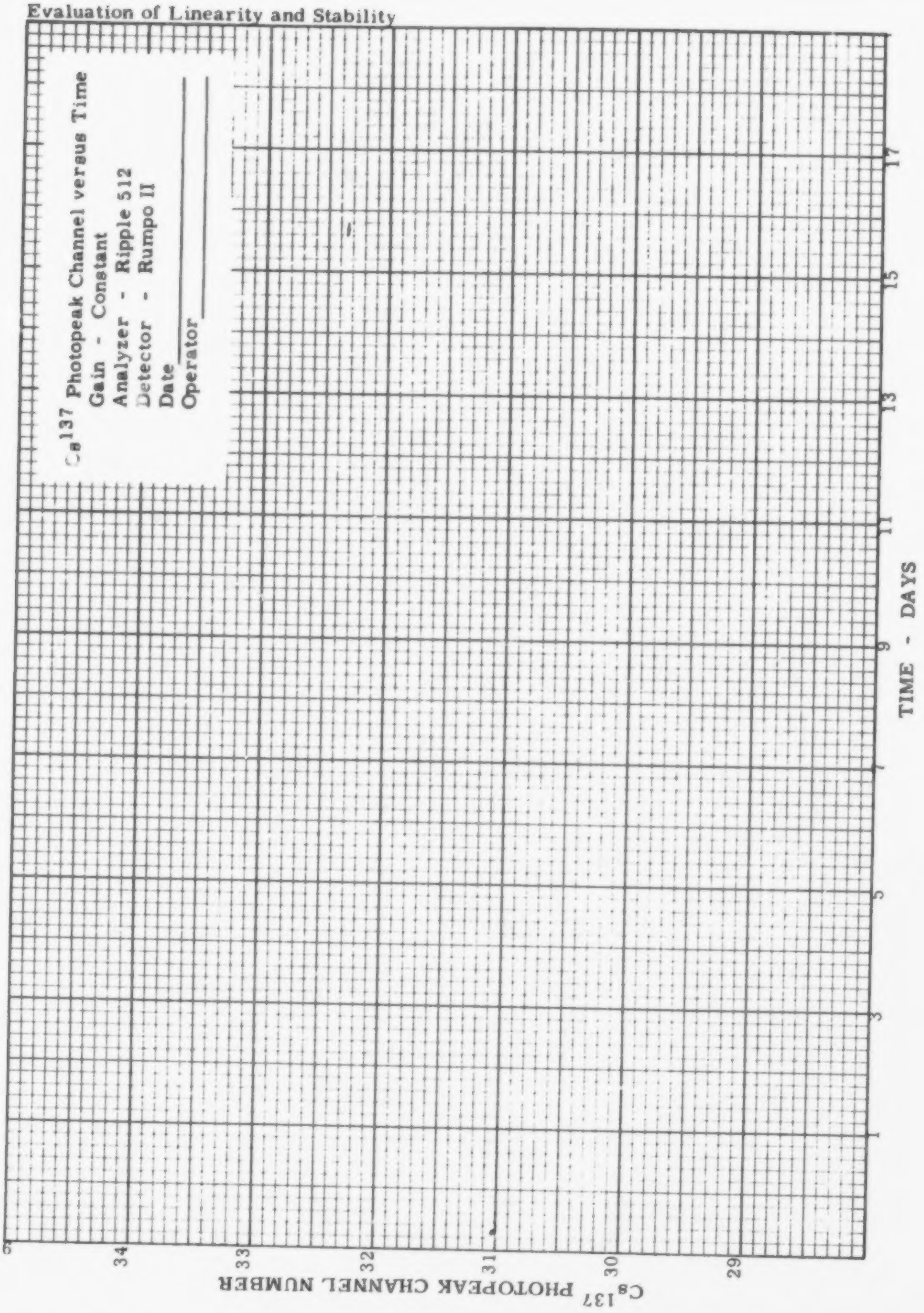
Channel coefficient: Approximately 20 Kev/channel

Detector: RUMPO I

Purpose for Data: Drift Analysis

Time (days)	Peak Channel
1	33.2
3	33.0
5	32.7
7	32.5
9	32.0
11	31.7
13	31.4

Evaluation of Linearity and Stability



Training Publication No. 366n

Efficiency Calibration

(Problem Session)

I. INTRODUCTION

This problem session is designed to give the trainee experience in calculating the detection efficiencies of the spectrometer for several radionuclides.

II. STATEMENT OF PROBLEM

Counting data from four nuclide standards, I^{131}, Cs^{137}, Ba-La^{140}, K^{40}, have been obtained. From this data it is required to calculate the total peak, total photopeak, and gamma photopeak efficiencies for each of the four nuclides. Efficiencies for the various nuclides are necessary to relate count rate to activity concentration.

III. PROCEDURE

From the attached data, calculate the total peak, total photopeak, and gamma photopeak efficiencies for each nuclide.

Record all data in Table 1.

Table 1. Calibration data sheet

Nuclide	Energy (Mev)	dpm Added	Peak Channel	Photopeak Channels	Gamma Abund.	Count Rate		Peak Eff.	Photo-peak Eff.	Gamma Eff.
						Peak	Photopeak			
K^{40}	1.46	188400	73	70–76						
I^{131}	0.364	12170	18	16–20						
Cs^{137}	0.663	11690	33	30–36						
Ba-La^{140}	0.489	24600	24	22–28						
	1.60	24600	80	76–85						

K^{40} STANDARD
1/16/64

TIME: 100 min.
ARRANGEMENT: Class A
VOLUME: 3.5 liters
ACTIVITY: 188400 dpm

20 Kev/channel
Net counts per minute
RUMPO I

Channel No.	Count	Channel No.	Count
06	70.82	41	9.49
07	61.40	42	9.30
08	56.46	43	10.01
09	48.57	44	9.69
10	41.81	45	9.80
11	41.67	46	10.70
12	43.05	47	11.43
13	40.64	48	11.39
14	34.01	49	11.58
15	29.41	50	10.84
16	24.40	51	11.67
17	22.24	52	10.90
18	19.75	53	12.10
19	18.29	54	11.52
20	17.10	55	11.36
21	15.27	56	12.40
22	14.95	57	12.40
23	13.50	58	12.78
24	11.97	59	11.81
25	12.85	60	11.29
26	12.52	61	8.50
27	11.75	62	7.67
28	11.16	63	6.18
29	10.29	64	5.04
30	10.52	65	4.33
31	10.93	66	3.54
32	10.04	67	4.49
33	9.51	68	6.24
34	10.14	69	11.10
35	9.87	70	19.60
36	10.61	71	30.46
37	9.95	72	40.58
38	9.70	73	43.92
39	9.86	74	37.06
40	9.36	75	26.71

K^{40} STANDARD--Continued

Channel No.	Count	Channel No.	Count
76	16.88	88	0
77	10.03	89	0
78	5.73	90	0
79	3.18	91	0
80	1.68	92	0
81	1.01	93	0
82	0.58	94	0
83	0.44	95	0
84	0.23	96	0
85	0.17	97	0
86	0.14	98	0
87	0	99	0

I^{131} STANDARD
1/16/64

TIME: 100 min.
ARRANGEMENT: Class A
VOLUME: 3.5 liters
ACTIVITY: 12,170 dpm

20 Kev/channel
Net counts per minute
RUMPO I

Channel No.	Count	Channel No.	Count
06	79.40	25	1.34
07	82.50	26	1.23
08	91.86	27	0.71
09	84.63	28	0.90
10	63.23	29	1.87
11	47.47	30	3.87
12	37.21	31	5.89
13	37.40	32	6.38
14	42.87	33	4.25
15	36.28	34	2.05
16	43.83	35	1.87
17	106.66	36	1.40
18	161.13	37	1.49
19	104.52	38	0.51
20	31.14	39	0.28
21	6.45	40	0
22	2.71	41	0
23	1.59	42	0
24	1.23		

Cs^{137} STANDARD
1/17/64

TIME: 100
ARRANGEMENT: Class A
VOLUME: 3.5 liters
ACTIVITY: 11690 dpm

20 Kev/channel
Net counts per minute
RUMPO I

Channel No.	Count	Channel No.	Count
06	40.77	24	9.04
07	40.08	25	6.88
08	38.05	26	7.11
09	44.08	27	6.03
10	43.79	28	6.14
11	36.51	29	7.47
12	31.49	30	14.56
13	26.26	31	34.35
14	23.84	32	63.05
15	20.83	33	67.05
16	19.99	34	50.14
17	18.92	35	23.51
18	16.69	36	8.32
19	16.66	37	2.84
20	16.50	38	1.09
21	15.62	39	0.41
22	13.86	40	0.17
23	11.69	41	0.10

Ba^{140} - La^{140} STANDARD
1/17/64

TIME: 100 min.
ARRANGEMENT: Class A
VOLUME: 3.5 liters
ACTIVITY: 24600

20 Kev/channel
Net counts per minute
RUMPO I

Channel No.	Count	Channel No.	Count
06	258.25	10	223.05
07	262.59	11	199.54
08	285.11	12	178.82
09	269.98	13	159.98

Ba^{140} - La^{140} STANDARD--Continued

Channel No.	Count	Channel No.	Count
14	147.34	57	17.06
15	157.34	58	16.31
16	183.76	59	16.27
17	158.34	60	16.35
18	99.62	61	16.23
19	72.45	62	17.34
20	72.30	63	17.35
21	80.86	64	17.12
22	92.04	65	16.83
23	121.63	66	16.35
24	162.54	67	14.93
25	161.81	68	13.23
26	132.52	69	11.23
27	105.24	70	9.97
28	75.17	71	8.75
29	49.68	72	8.22
30	35.77	73	7.48
31	29.78	74	7.86
32	27.11	75	9.24
33	25.08	76	15.01
34	23.59	77	22.71
35	24.03	78	35.41
36	24.54	79	45.47
37	27.33	80	52.16
38	32.45	81	50.74
39	40.31	82	41.45
40	49.83	83	29.36
41	53.76	84	19.50
42	48.81	85	12.12
43	41.35	86	9.39
44	33.43	87	7.02
45	29.64	88	5.50
46	29.77	89	4.33
47	27.14	90	4.24
48	22.91	91	3.73
49	18.02	92	3.92
50	16.45	93	3.33
51	16.37	94	2.98
52	14.41	95	2.95
53	15.24	96	2.51
54	16.73	97	2.58
55	16.26	98	2.55
56	17.98	99	2.77

Statistical Evaluation of Gamma Spectra

Problem Session

I. INTRODUCTION

This problem session will provide practice in applying some basic concepts of nuclear counting statistics to gamma spectroscopy.

II. STATEMENT OF PROBLEM

Counting data for two samples have been obtained. From this data and the RHINO I Gamma Spectrum Catalog determine the appropriate values and errors at the 95% confidence level.

III. SINGLE NUCLIDE COUNTING PROBLEM

Given:

An analytical determination uses barium-133 as a tracer to determine the chemical yields of the procedure. The analyst using RHINO I analyzer decides to use the peak channel to minimize background. He collects 1076 counts in 2 minutes from his barium-133 tracer spike.

Find:

A. The counting error associated with the net counting rate both in cpm and in % relative error. Use a background counting time of 2 minutes and Class B background.

B. The chemical yield of the analysis with its error. The added dpm is 80,000 ± 5% relative error at 95% C.L.

C. The optimum number of channels to minimize relative statistical error. Calculate the relative error using the optimum number of channels involved from the Ba^{133} standard in RHINO I catalog and compare it to peak channel relative error for the standard.

IV. MINIMUM DETECTABLE ACTIVITY PROBLEM

Given:

Sample = Cs^{137}

Sample size = 3.5 liters

Chemical Yield = 75%

Background Counting Time (Class A) No. II = 100 minutes

Sample Counting Time = 100 minutes

Total Photopeak Efficiency = 0.0199

Find:

A. The M.D.A. and the relative error associated with it at the 95% Confidence Level.

B. The lowest count rate when a 25% precision at 95% Confidence Level is desired.

C. The lowest activity concentration using the N_s found in B.

D. The lowest activity concentration for gross gamma counting (ch 6 to ∞) where the precision at the 95% Confidence Level is 25%.

SINGLE NUCLIDE COUNTING PROBLEM

WORKSHEET

A. $Ba^{133}N_s$ = ________________ cpm ± ________________ cpm (95%)

= ________________ cpm ± ________________ % "

B. Chemical Yield = 0.________________ ± 0.________ "

C. Optimum Channels = ________________________________

Standard Optimum Channels Relative Error = ________ % "

Standard Single Channel Relative Error = ________ % "

MINIMUM DETECTABLE ACTIVITY PROBLEM

WORKSHEET

A. MDA = ______________ cpm % error (95% C.L.) = _________

B. ACT. = ______________ cpm

C. ACT. CONC. = ______________ pc/1

D. ACT. CONC. = ______________ pc/1

Training Publication No. 379n

Qualitative Gamma Analysis - Radionuclide Identification by Energy and Half-Life

(Problem Session)

I. INTRODUCTION

This problem session is designed to give the trainee experience in the procedure of radionuclide identification by energy and half-life from gamma-ray spectra.

II. STATEMENT OF PROBLEM

A sample of water was collected from a radiochemistry drainage system. The sample was counted on a multichannel gamma spectrometer at four different times. The graphical spectra and the data are included on the following pages. It is required to determine the radionuclide composition of the sample.

III. THE PROCEDURE

A suggested procedure for the solution of the problem is as follows: (Table I may be used as a guide for the procedure)

A. Determine energy of each characteristic peak.

B. List nuclides and their associated half-lives which emit gamma photons of energies within a range of plus or minus one channel from determined peaks. Use table entitled "Radionuclides Arranged by Gamma Ray Energies".

C. Determine half-lives for peaks in composite spectra.

1. Draw in Compton background on each of the four spectra.

2. Determine peak cpm minus Compton cpm for each energy peak. This gives an estimate of relative activity.

3. Plot relative activity vs time of count on semilog paper.

4. Determine half-life of peak from semilog plot.

D. Eliminate possibilities from half-life determination.

1. Compare half-lives of possibilities determined in Step B with half-lives determined from Step C.

2. Reject those nuclides whose half-lives disagree badly.

E. Make final estimate of nuclides in sample. Use "Gamma Spectrum Catalog" to compare remaining possibilities with sample composite spectra.

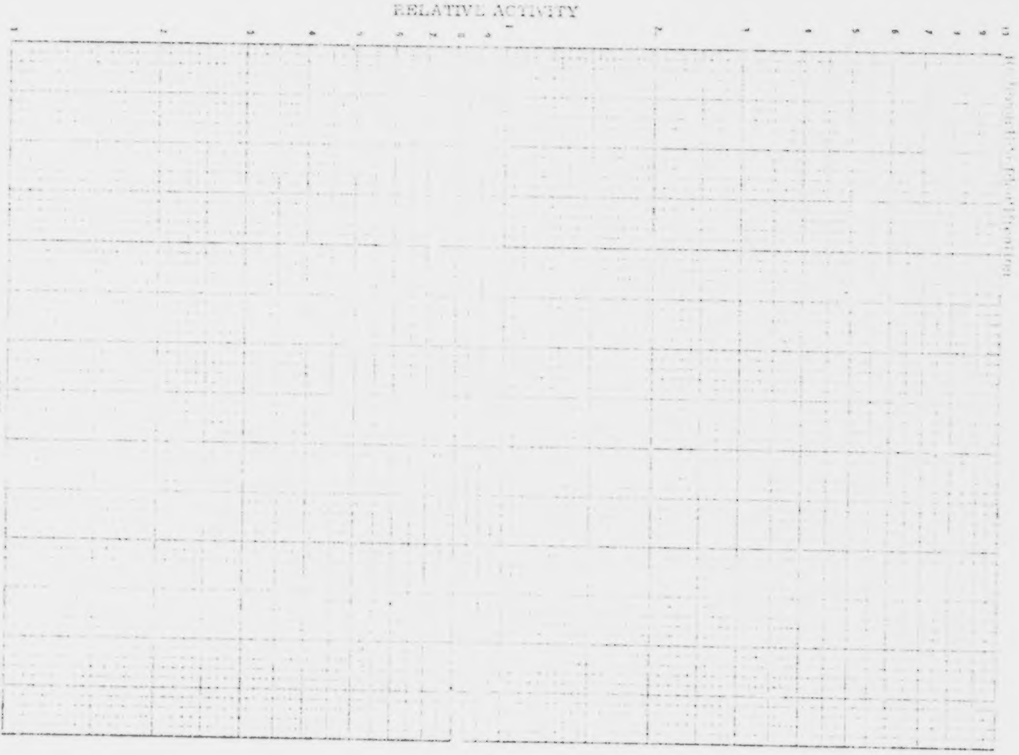
Table 1

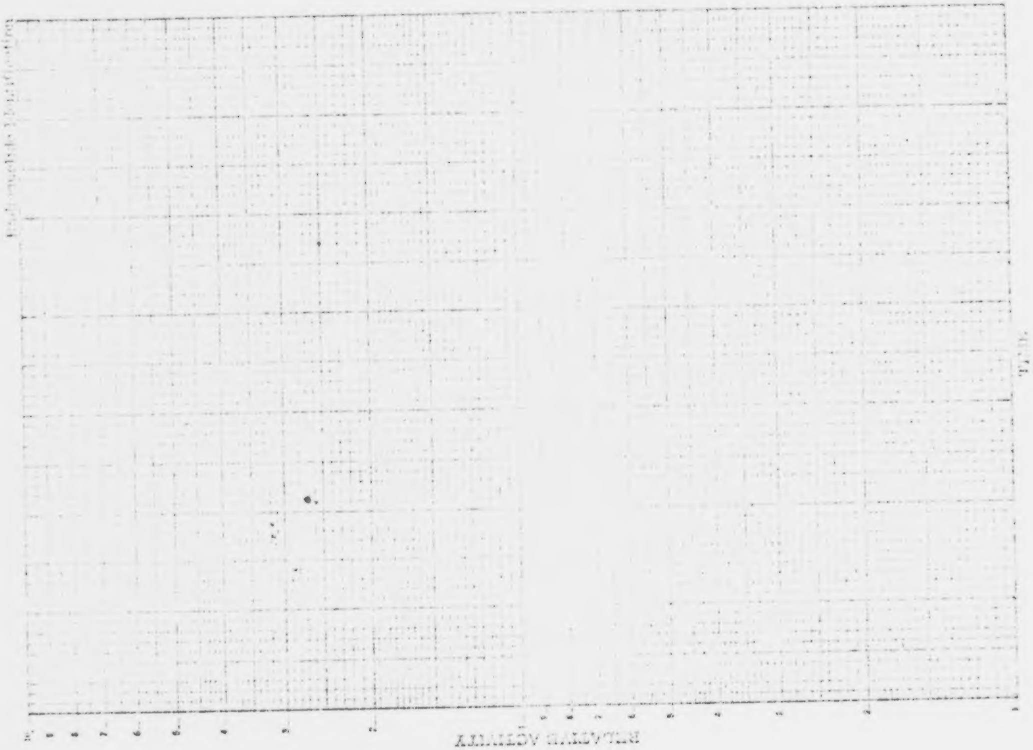
Determined Peak Energy	Determined Half-Life	Possible Nuclides	Actual Energies of Possible Nuclides	Actual Half-Lives	Confirmed Nuclides

Table 1 (cont.)

Determined Peak Energy	Determined Half-Life	Possible Nuclides	Actual Energies of Possible Nuclides	Actual Half-Lives	Confirmed Nuclides

CALCULATIONS


RELATIVE ACTIVITY
TIME


Radionuclide Identification
RELATIVE ACTIVITY
TIME

RELATIVE ACTIVITY

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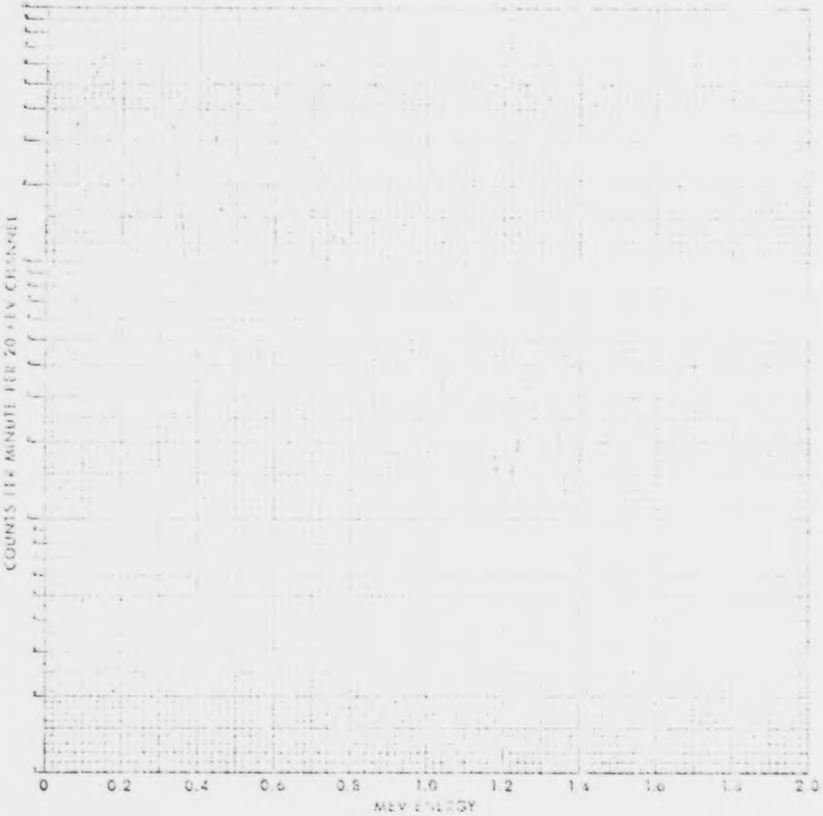
COUNTING ARRANGEMENT [illegible]

COUNTING TIME [illegible]

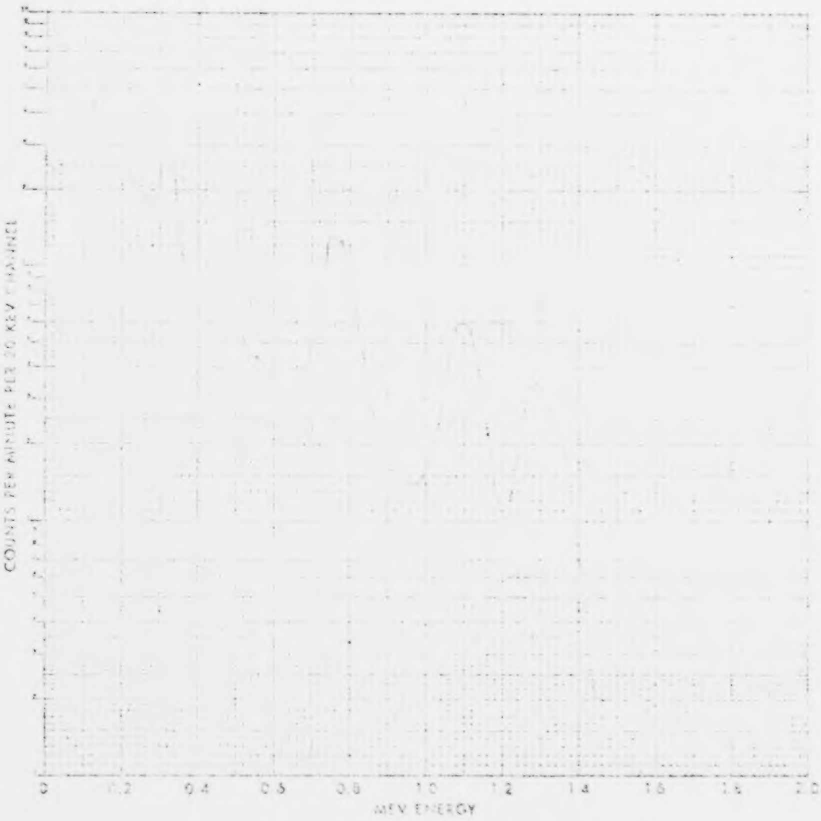
CPM		CPM	

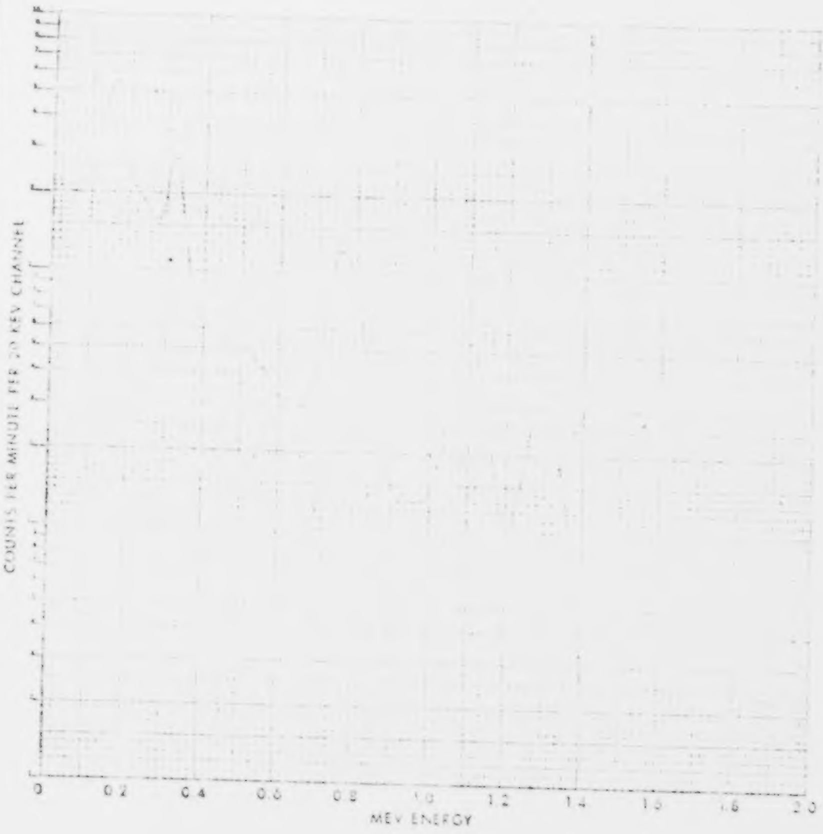


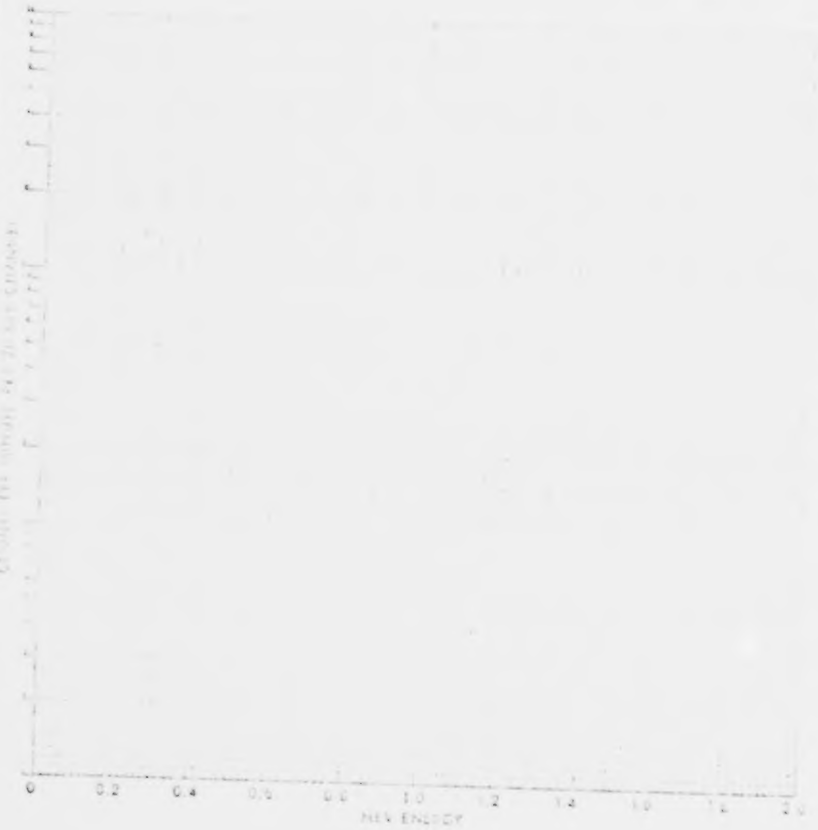
SAMPLE [illegible]

COUNTING ARRANGEMENT [illegible]

COUNTS [illegible]




COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY


COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY

Qualitative Gamma Analysis
Chemical Separation

Problem Session

I. INTRODUCTION

The purpose of this problem session is to illustrate the significance and use of chemical separation in the identification of radionuclides in a water sample.

II. STATEMENT OF PROBLEM

A sample of water was collected near a radiochemistry laboratory drainage system. Gross analysis indicated a concentration of about 700 pc/l as cesium-137 apparent beta activity, and no gross alpha. Aliquots of the sample were taken for strontium-90 analysis, radium-226 analysis, and gamma spectral analysis. The gamma spectrum of the original sample was too complex for definite identification of all the radionuclides in the sample. Therefore, carriers isotopic to the suspected radionuclides were added, and a qualitative chemical separation was performed, separating the radionuclides into the following six groups:

A. Added Carriers - I, Hg, Ru, Sb, Ce, Cr, Fe, Zr, Nb, Co, Mn, Zn, Sr, Ba, Na, Cs

1. Silver Group
2. Acid Sulfide Group
3. Hydroxide Group
4. Basic Sulfide Group
5. Alkaline Earth Group
6. Soluble Group

The original sample, each chemical group, and the net sample after each separation were counted on a gamma spectrometer. Both the graphical spectra and the data are included on the following sheets. It is required to determine the radionuclide composition of the sample.

III. PROCEDURE

A suggested procedure for the solution of the problem is as follows: (Table 1 may be used as a guide for the procedure).

A. For each spectrum determine the energy of each characteristics peak.

B. List nuclides which emit gamma photons of energies within a range of plus or minus one channel from determined peaks. Use table entitled "Radionuclides Arranged by Gamma-Ray Energies".

C. From knowledge of the energies of identified peaks and the chemistry of the nuclides, make qualitative identification of the radionuclides present in the original sample.

Table 1

Determined peak energies	Possible nuclides	Actual energies of possible nuclides	Nuclides in chemical group	Confirmed nuclides
NET SAMPLE SPECTRUM				
SILVER GROUP				
SAMPLE SPECTRUM MINUS SILVER GROUP				
ACID SULFIDE GROUP				

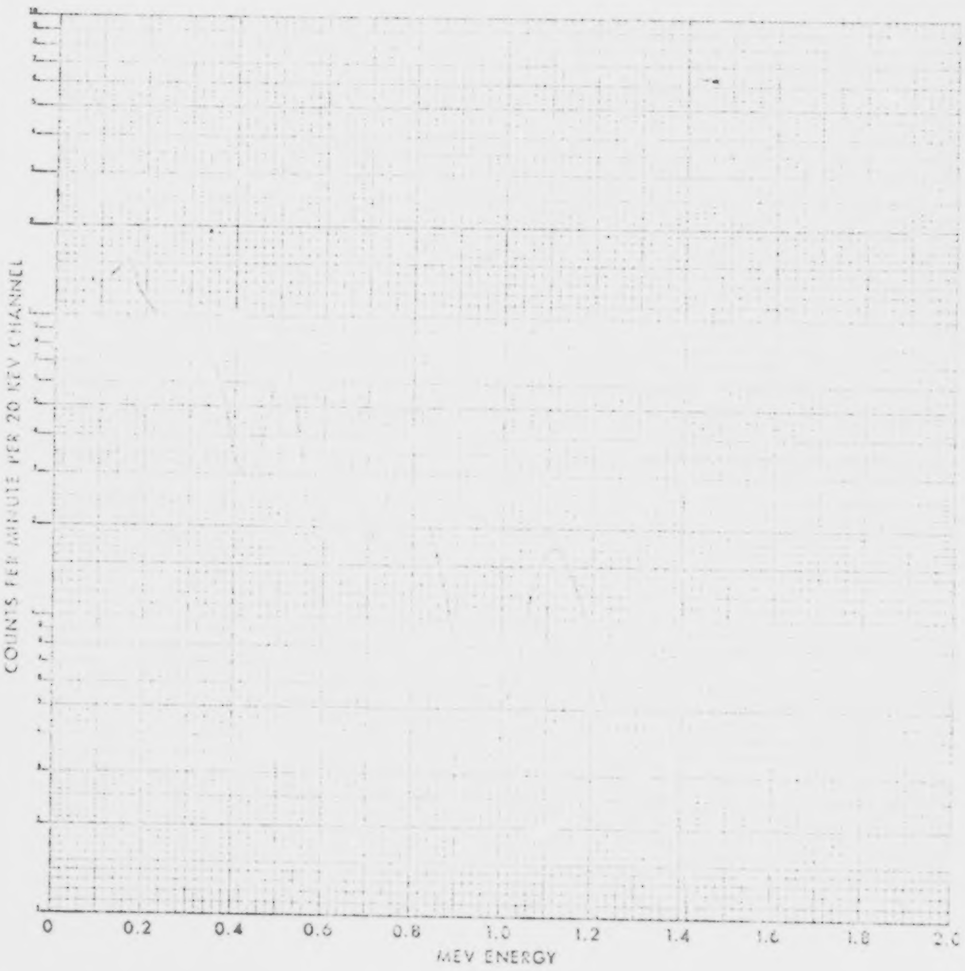
Table 1

Determined peak energies	Possible nuclides	Actual energies of possible nuclides	Nuclides in chemical group	Confirmed nuclides
SAMPLE SPECTRUM MINUS SILVER AND ACID SULFIDE GROUPS				
HYDROXIDE GROUP				
SAMPLE SPECTRUM MINUS SILVER, ACID SULFIDE, AND HYDROXIDE GROUPS				
BASIC SULFIDE GROUP				

Table 1

Determined peak energies	Possible nuclides	Actual energies of possible nuclides	Nuclides in chemical group	Confirmed nuclides
SAMPLE SPECTRUM MINUS FIRST FOUR GROUPS				
ALKALINE EARTH GROUP				
SOLUBLE GROUP				

SAMPLE [illegible]
ACTIVITY [illegible]
COUNTING ARRANGEMENT [illegible]
SAMPLE VOLUME [illegible]
DETECTOR [illegible] ANALYZER [illegible]
DATE [illegible]
COUNTING TIME [illegible]



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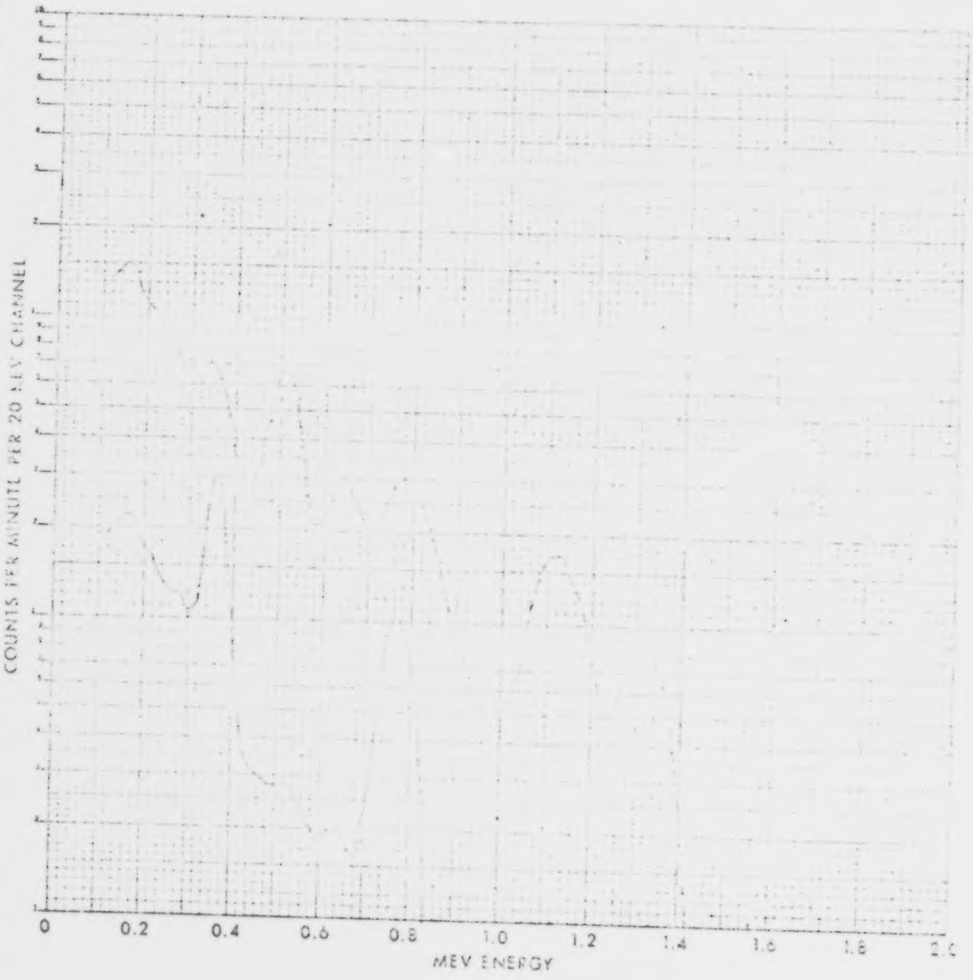
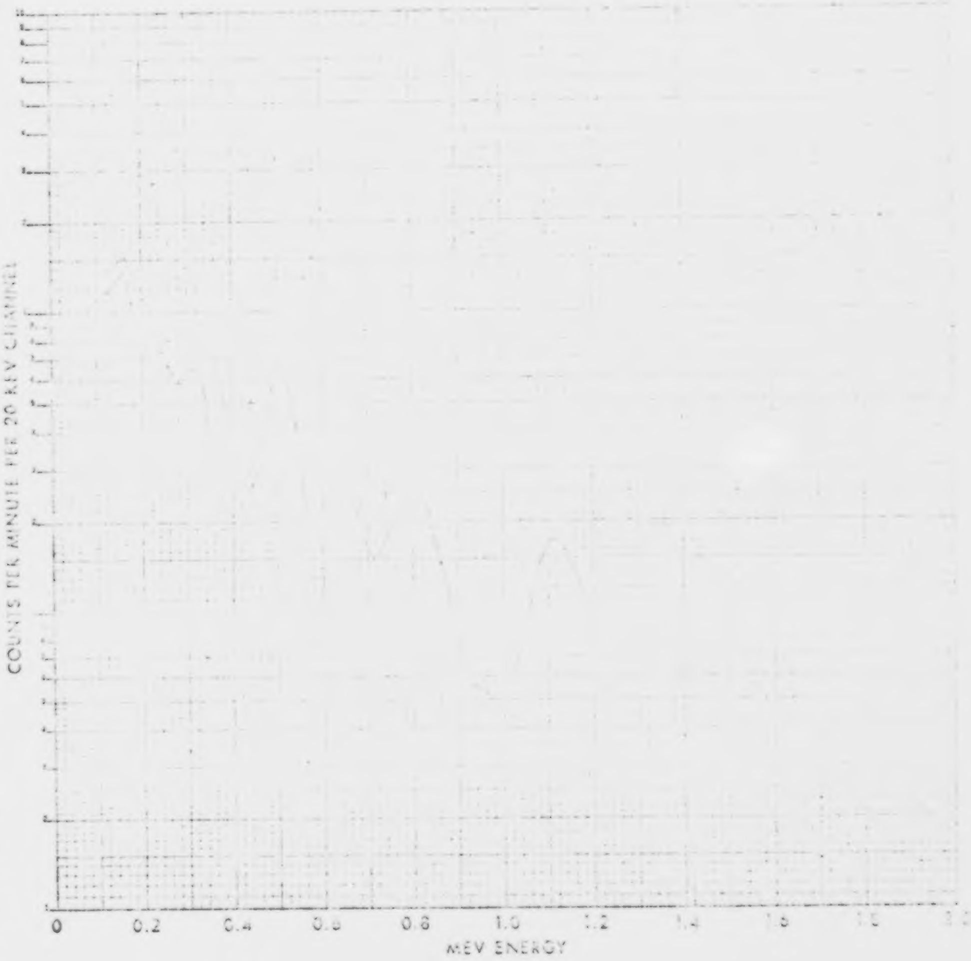
ACTIVITY [illegible]
COUNTING ARRANGEMENT [illegible]
SAMPLE VOLUME [illegible]
DETECTOR [illegible] ANALYZER [illegible]
DATE [illegible]
COUNTING TIME [illegible]

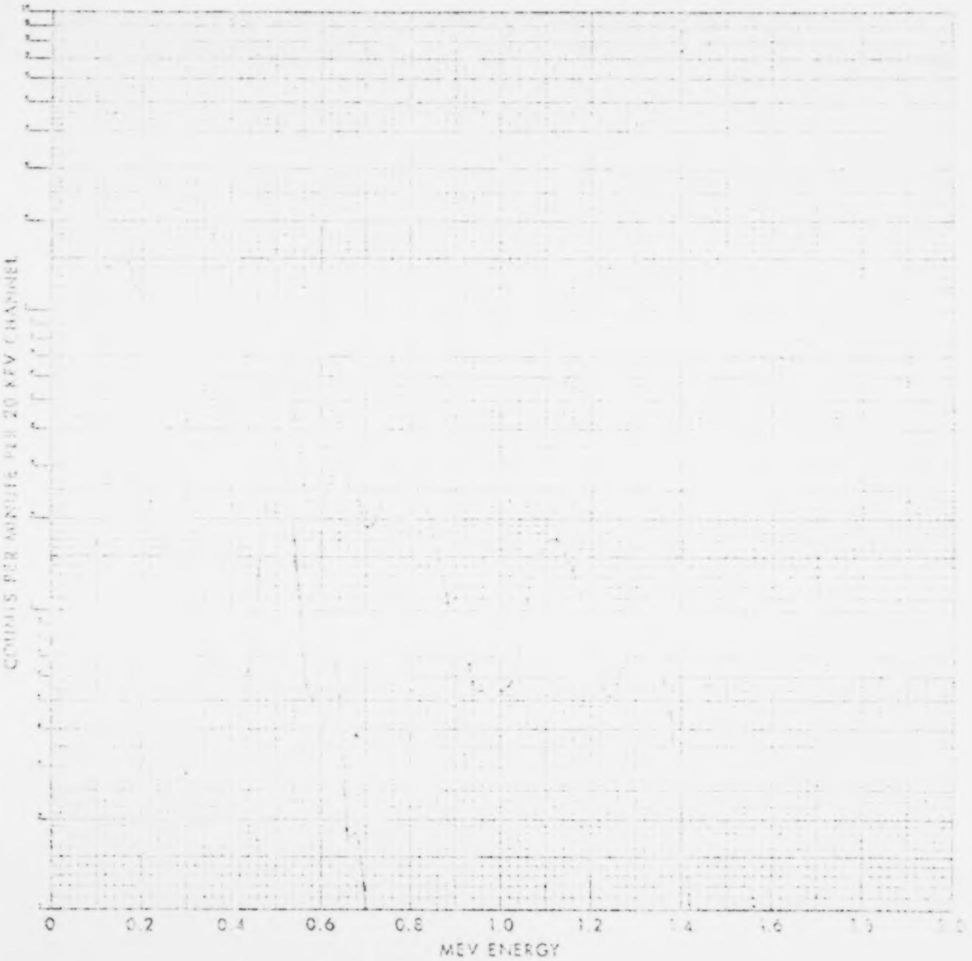
CPM	CPM
[illegible]	[illegible]



SAMPLE
ACTIVITY
COUNTING ARRANGEMENT
SAMPLE VOLUME
DETECTOR ANALYZER
DATE
COUNTING TIME



COUNTING TIME



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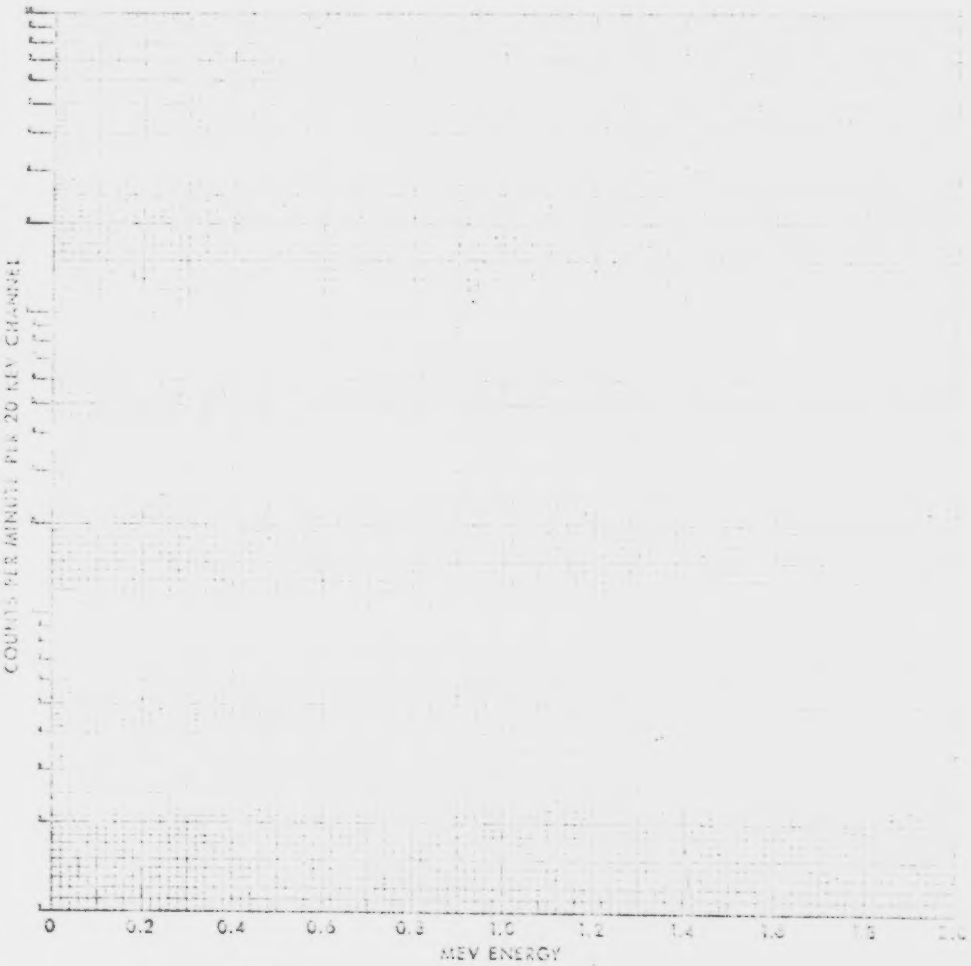
COUNTING ARRANGEMENT [illegible]

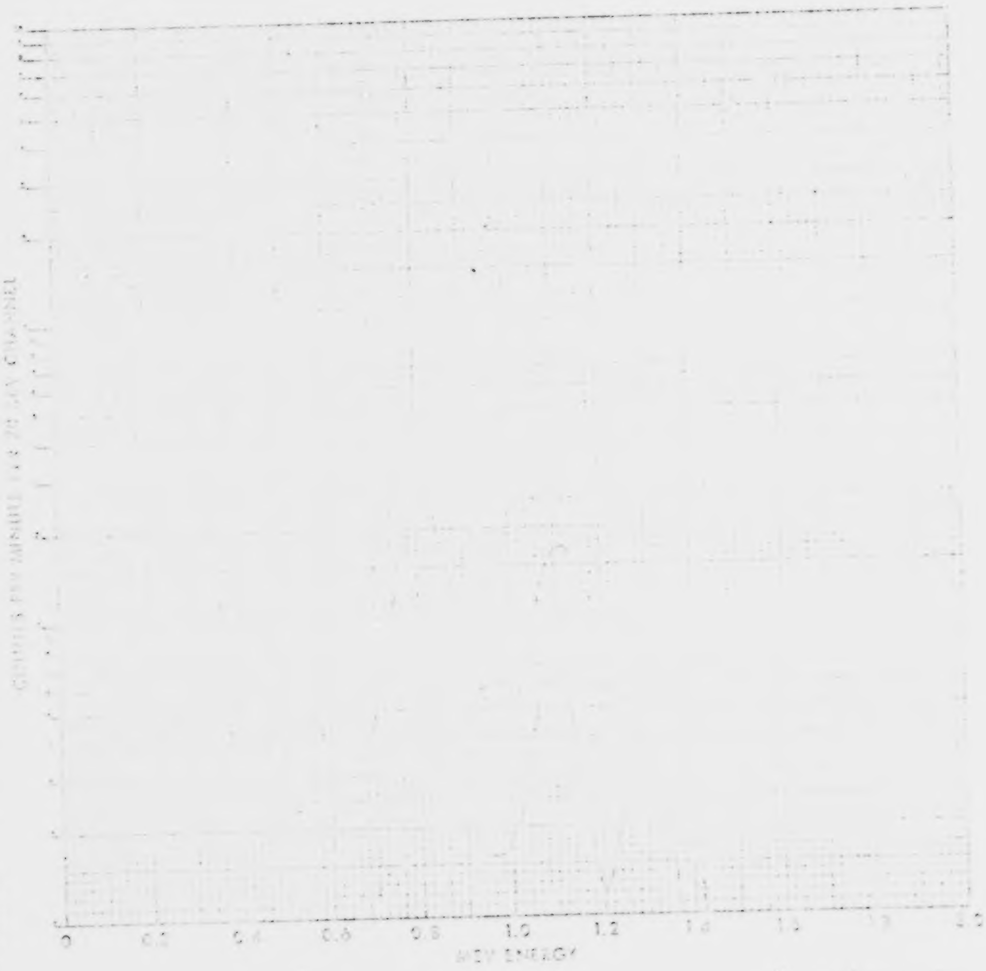
SAMPLE VOLUME [illegible]

DETECTOR [illegible] ANALYZER 400-V

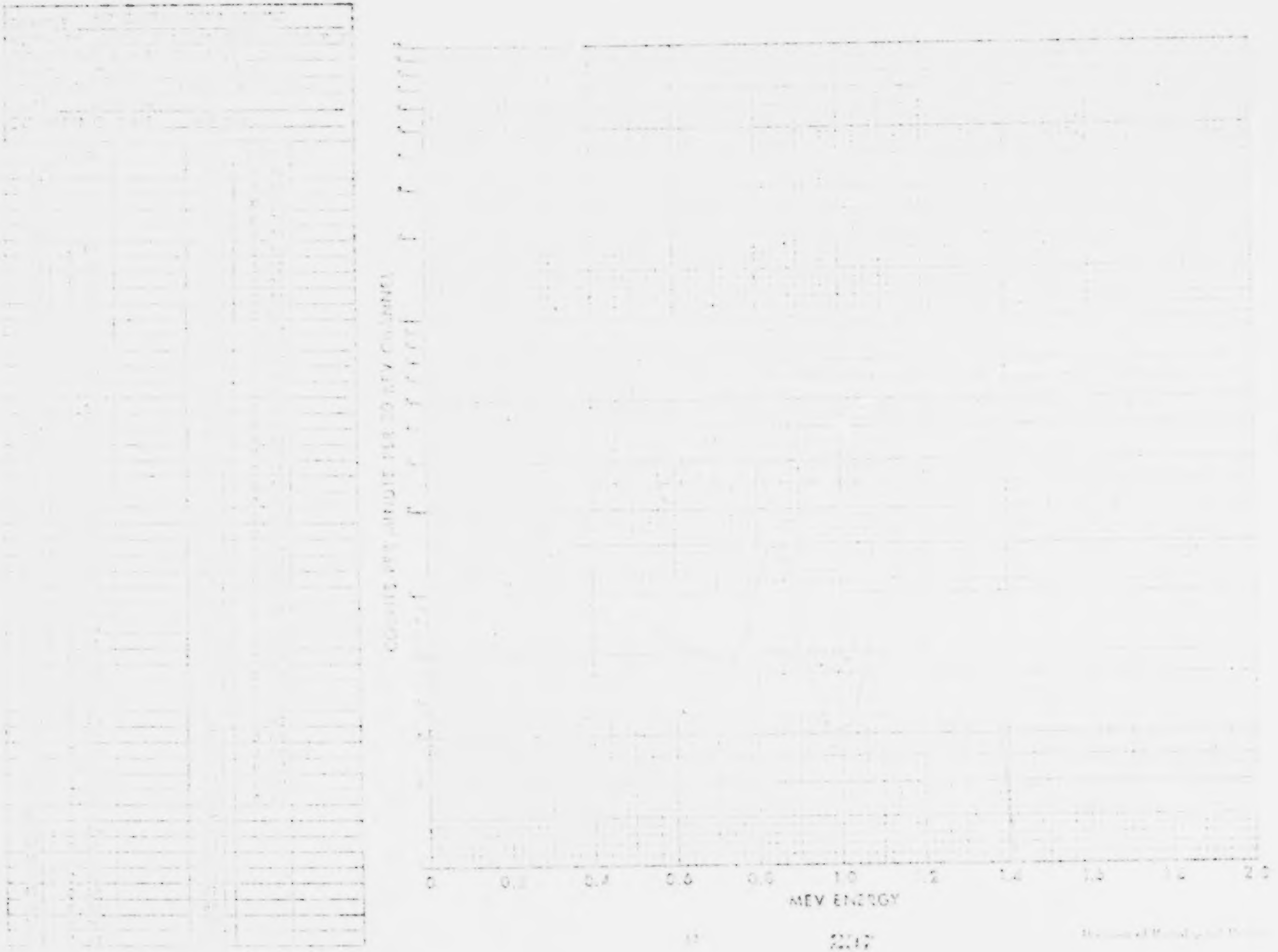
DATE [illegible]

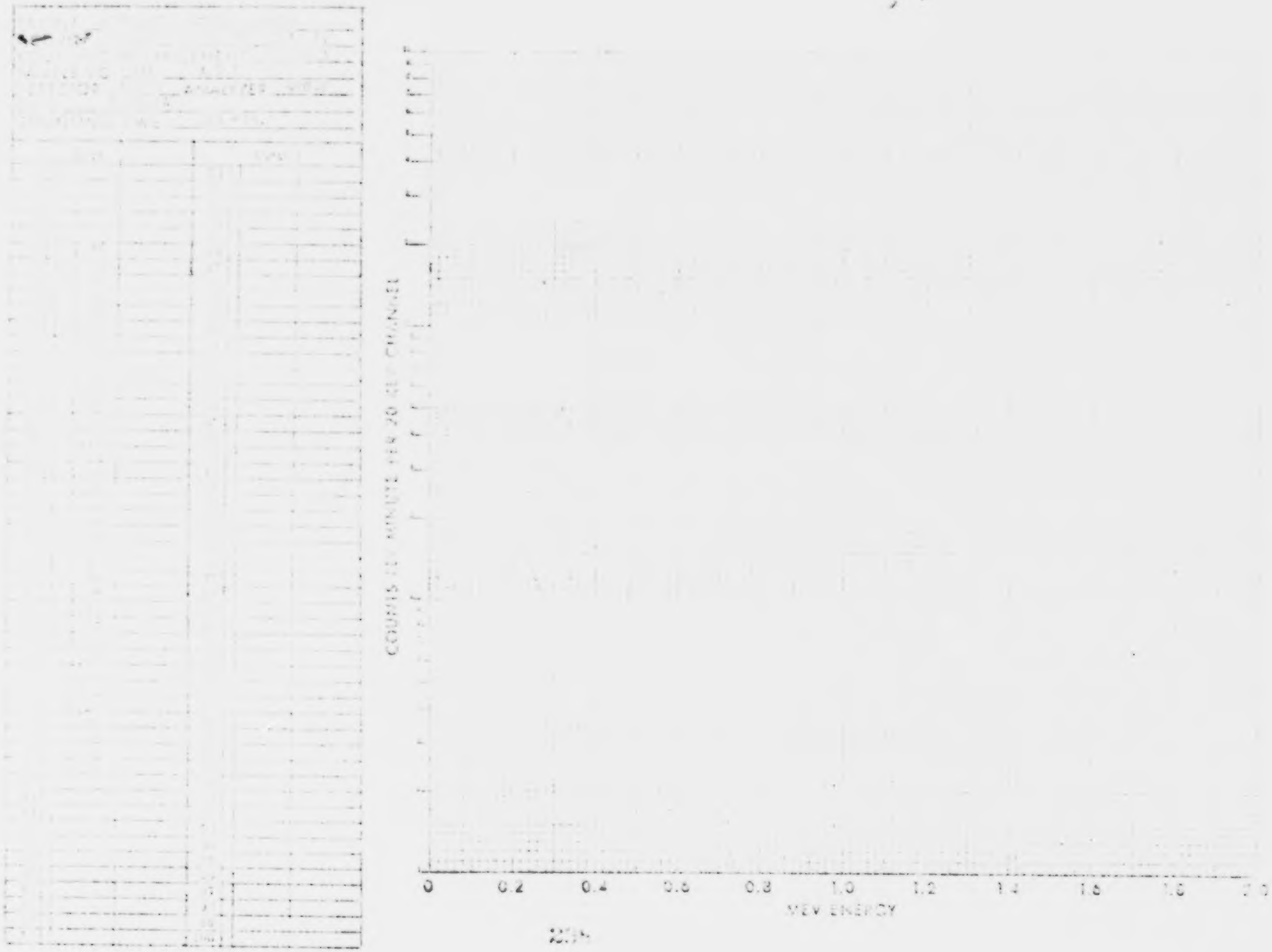
COUNTING TIME [illegible]




COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY

SAMPLE
LOCATION
COUNTING ARRANGEMENT
SAMPLE VOLUME
DETECTOR
DATE
COUNTING TIME
ANALYZER
COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY

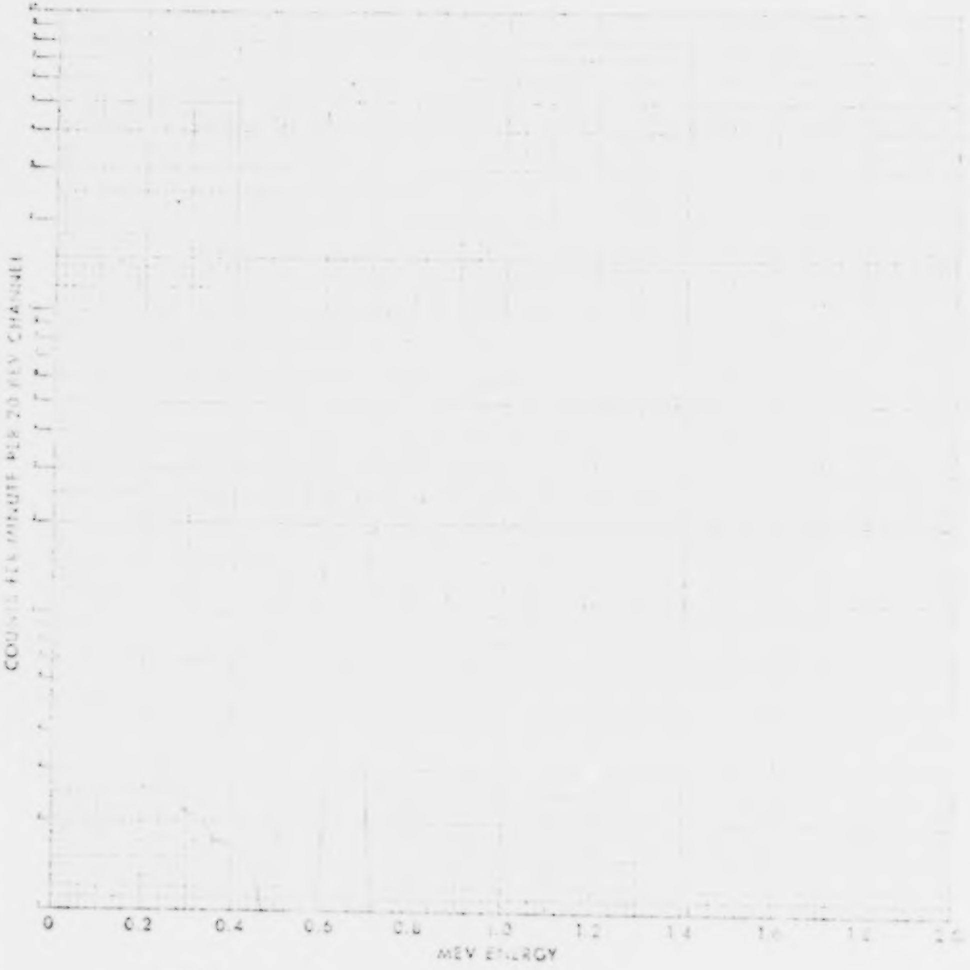

COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY


COUNTS PER MINUTE PER 20 KEV CHANNEL
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
MEV ENERGY
ANALYZER

SAMPLE [illegible]

[illegible]

COUNTING TIME [illegible]



Analysis of Milk Samples
Compton Continuum and Matrix Methods

LABORATORY

A milk sample was obtained from a 14 yr. old Jersey cow. A 3.5 liter portion of the sample was analyzed by gamma spectroscopy yielding the spectrum shown in Figure 1. It is suspected that the milk contains traces of Ba^{140} - La^{140}, I^{131}, Cs^{137}, K^{40}.

A. Confirm radionuclide composition through peak identification.

B. Estimate the radionuclide concentrations in pc/l by subtracting the Compton Continuum background from the total photopeak areas. Use the following selected channels and efficiencies.

Total photopeak efficiencies from Compton Continuum Subtraction Method.

I^{131}	- .0314	(Channels 16-20)
Ba^{140} - La^{140}	- .02033	(Channels 22-28)
Cs^{137}	- .0191	(Channels 30-36)
K^{40}	- .000979	(Channels 70-76)

C. Determine the radionuclide concentration in pc/l by the simultaneous equation method. Use efficiencies from the "Gamma Spectrum Catalogue - RHINO I".

Solutions to Simultaneous equations.

$I_i = \underline{\quad\quad} N_i \underline{\quad\quad} N_b \underline{\quad\quad} N_c \underline{\quad\quad} N_k$

$B_b = \underline{\quad\quad} N_i \underline{\quad\quad} N_b \underline{\quad\quad} N_c \underline{\quad\quad} N_k$

$C_c = \underline{\quad\quad} N_i \underline{\quad\quad} N_b \underline{\quad\quad} N_c \underline{\quad\quad} N_k$

$K_k = \underline{\quad\quad} N_i \underline{\quad\quad} N_b \underline{\quad\quad} N_c \underline{\quad\quad} N_k$

$$pc/l = \frac{cpm}{eff.\ (Vol)\ (2.22\ dpm/pc)}$$

WORK SHEET

COMPTON CONTINUM SUBTRACTION

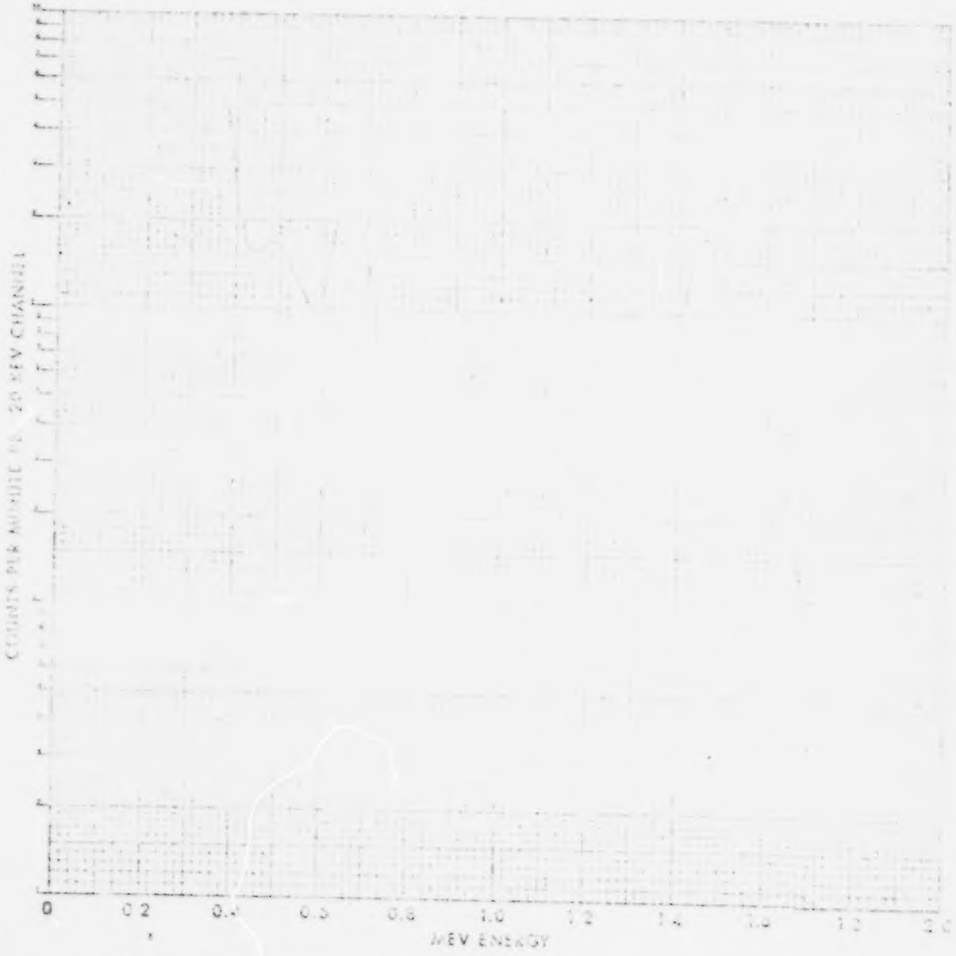
PHOTOPEAK	CHANNEL NO.	PEAK AREA	COMPTON BACKGROUND	ACTIVITY cpm
I^{131}	16			
	17			
	18			
	19			
	20			
	TOTAL			
Ba^{140}-La^{140}	22			
	23			
	24			
	25			
	26			
	27			
	28			
	TOTAL			
Cs^{137}	30			
	31			
	32			
	33			
	34			
	35			
	36			
	TOTAL			
K^{40}	70			
	71			
	72			
	73			
	74			
	75			
	76			
	TOTAL			

RESULTS

QUANTITATIVE ANALYSIS OF MILK SAMPLE

RADIO-NUCLIDE	ACTUAL ACTIVITY	ACTIVITY BY COMP. SUB.	% ERROR	ACTIVITY BY MATRIX	% ERROR
I^{131}					
Ba^{140}-La^{140}					
Cs^{137}					
K^{40}					

SAMPLE [illegible]
ACTIVITY [illegible]
COUNTING ARRANGEMENT [illegible]
SAMPLE VOLUME [illegible]
DETECTOR [illegible] ANALYZER [illegible]
DATE [illegible]
COUNTING TIME [illegible]



Division of Radiological Health

Spectrum Stripping - Quantitative Analysis

(Problem Session)

I. INTRODUCTION

The purpose of this problem session is to provide practice in applying the basic concept of spectrum stripping to the quantitative analysis of a gamma spectrum. The spectra are hypothetical and are not intended to represent an actual analysis problem.

II. SAMPLE DATA

A 3.5 liter sample of Martian milk was obtained and counted on a gamma spectrometer. The resulting spectrum, see Figure 1, shows the sample to contain Bombium-158, Mullencium-24, and Ratsecium-66. It is desired to determine the activity concentration of the milk.

III. SPECTROMETER CALIBRATION

Channel Coefficient: 50 Kev per channel

Nuclide	Spectral Plot	Selected Photopeak Channels	Total Photopeak Efficiency	Energy
Bombium-158	Figure 2	5-7	7.2%	.30 Mev
Mullencium-24	Figure 3	9-11	5.9%	.50 Mev
Ratsecium-66	Figure 4	15-17	3.8%	.80 Mev

IV. PROCEDURE

1. Subtract the highest energy peak from sample spectrum by the following steps:

a. Determine the spectrum multiplier using selected photopeak channels.

*b. Multiply the count rate in Channel 1 of the standard spectrum by the spectrum multiplier.

*c. Subtract this adjusted count rate from the sample spectra count rate in Channel 1.

d. Proceed as in steps b and c (above) for all channels.

e. Plot the resulting subtraction spectrum.

2. "Strip from the resulting spectrum the next highest energy peak by the same method as given above.

3. The final subtraction spectrum should be identical in shape to the standard spectrum of the lowest energy peak. Confirm by "stripping" the final single peak.

4. Determine the net count rate of each photopeak which represents a nuclide.

5. Calculate the activity concentrations of each nuclide in pc/1.

* Steps 1b and 1c, when done on an electric calculator, are most efficiently performed by using negative multiplication to adjust the standard and then "adding" the sample spectrum to obtain the difference.

V. CALCULATION DATA

Nuclide	Spectrum Multiplier	Net Count Rate in Photopeak Channels cpm	Activity Concentration pc/1
Ratsecium-66			
Mullencium-24			
Bombium-158			

COUNTING DATA AND CALCULATION SHEET
Counts Per Minute

Channel Number	Sample	Rt^{66} Standard S.M.=__	First Subtraction Spectrum	Mu^{24} Standard S.M.=__	Second Subtraction Spectrum	Bo^{158} Standard S.M.=__	Third Subtraction Spectrum
01	63.90	420		380		320	
02	61.47	390		390		300	
03	54.46	350		360		230	
04	42.96	290		300		120	
05	55.77	280		390		390	
06	62.29	278		430		530	
07	53.17	276		360		370	
08	31.48	275		130		90	
09	45.81	270		450		0	
10	51.31	270		550		0	
11	42.83	260		410		0	
12	29.40	250		180		0	
13	17.94	230		0		0	
14	15.60	200		0		0	
15	34.32	440		0		0	
16	45.24	580		0		0	
17	31.98	410		0		0	
18	8.58	110		0		0	
19	0	0		0		0	

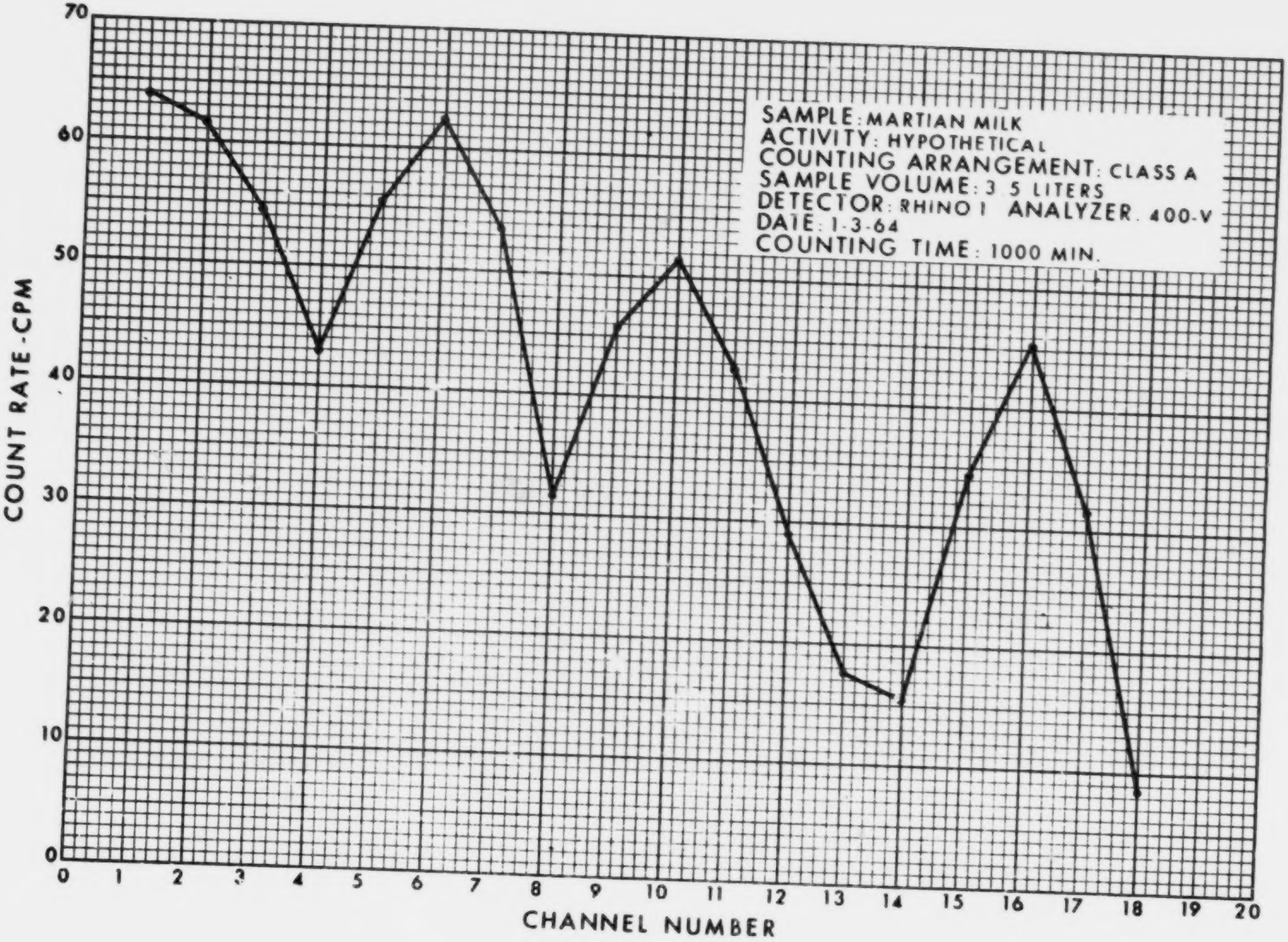

SAMPLE: MARTIAN MILK
ACTIVITY: HYPOTHETICAL
COUNTING ARRANGEMENT: CLASS A
SAMPLE VOLUME: 3 5 LITERS
DETECTOR: RHINO I ANALYZER. 400-V
DATE: 1-3-64
COUNTING TIME: 1000 MIN.
COUNT RATE - CPM
70
60
50
40
30
20
10
0
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
CHANNEL NUMBER

FIGURE 1

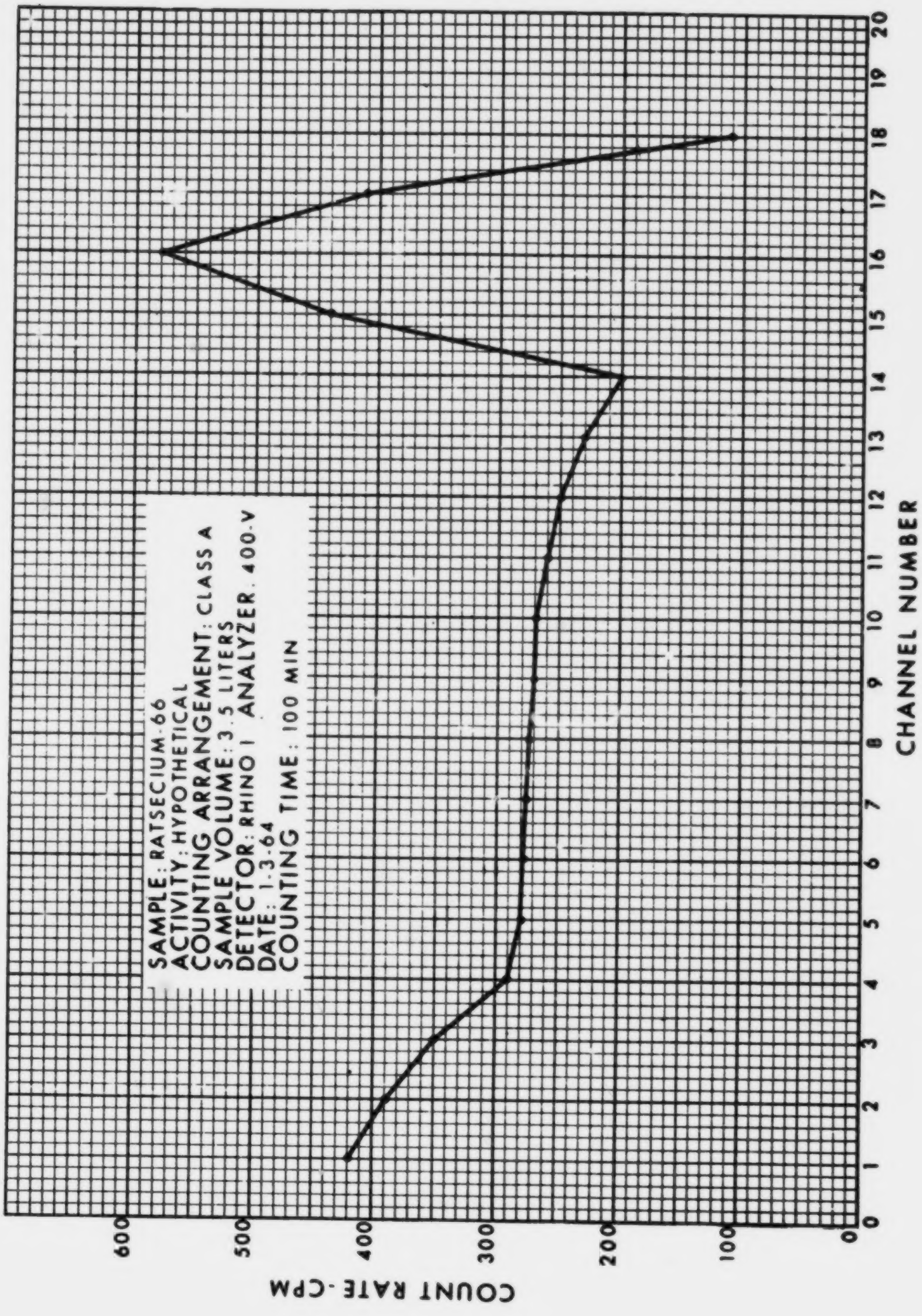
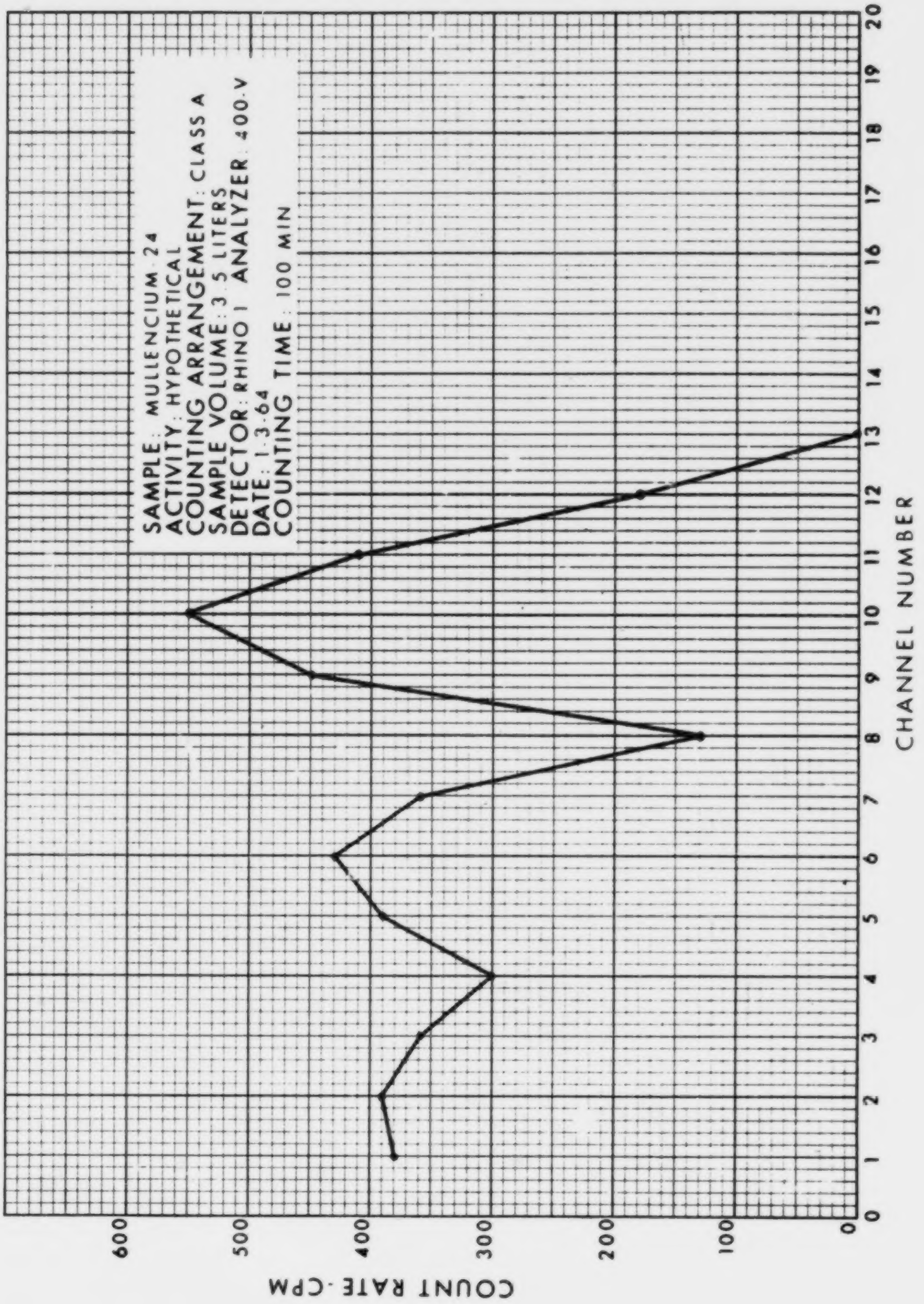

SAMPLE: RATSECIUM-66
ACTIVITY: HYPOTHETICAL
COUNTING ARRANGEMENT: CLASS A
SAMPLE VOLUME: 3.5 LITERS
DETECTOR: RHINO I ANALYZER. 400-V
DATE: 1-3-64
COUNTING TIME: 100 MIN
COUNT RATE-CPM
0
100
200
300
400
500
600
CHANNEL NUMBER
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20

FIGURE 2

308


SAMPLE: MULLENCIUM-24
ACTIVITY: HYPOTHETICAL
COUNTING ARRANGEMENT: CLASS A
SAMPLE VOLUME: 3.5 LITERS
DETECTOR: RHINO I ANALYZER: 400-V
DATE: 1-3-64
COUNTING TIME: 100 MIN
COUNT RATE - CPM
0
100
200
300
400
500
600
CHANNEL NUMBER
0
1
2
3
4
5
6
7
8
9
10
11
12
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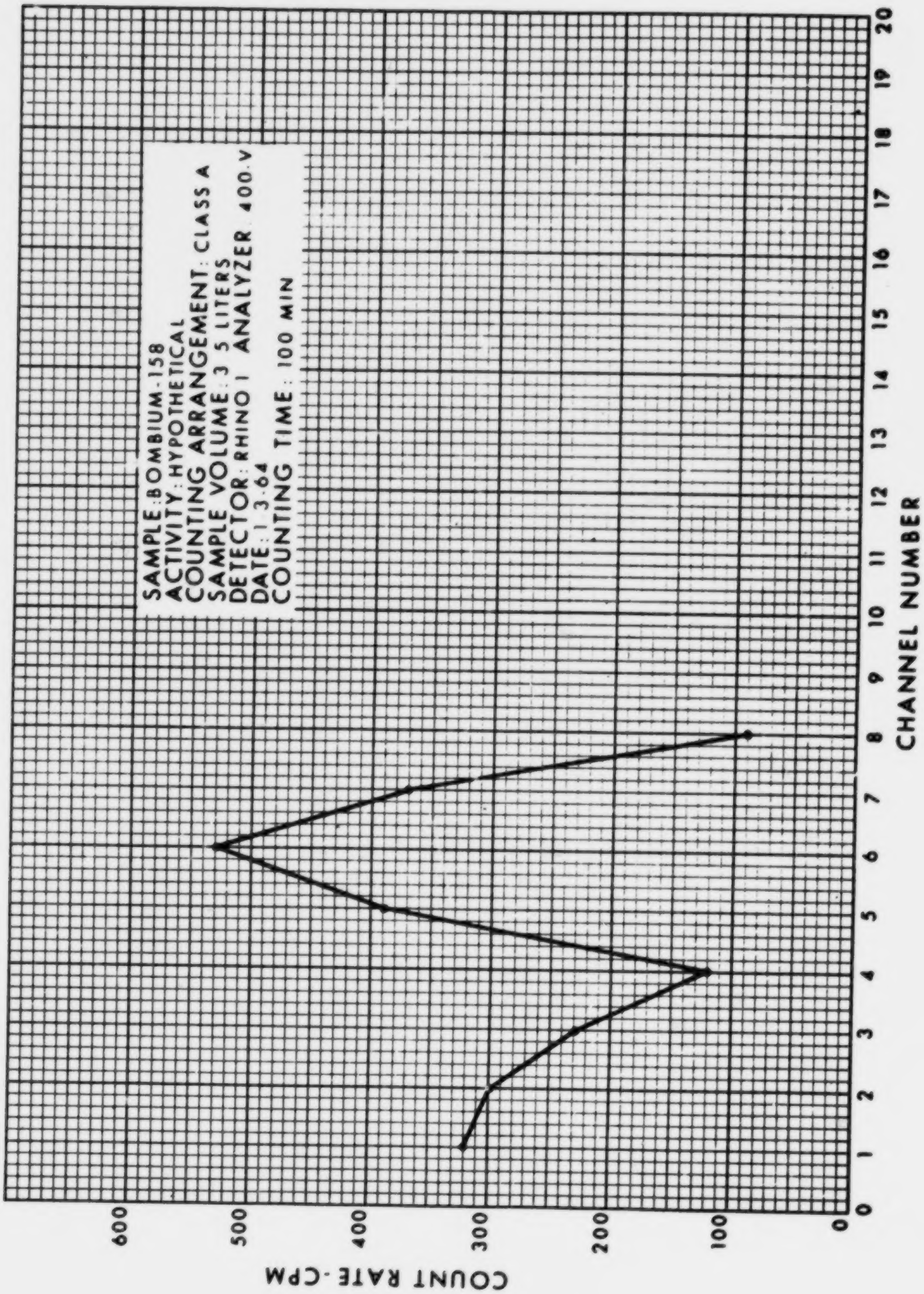
14
15
16
17
18
19
20

FIGURE 3


SAMPLE: BOMBIUM-158
ACTIVITY: HYPOTHETICAL
COUNTING ARRANGEMENT: CLASS A
SAMPLE VOLUME: 3.5 LITERS
DETECTOR: RHINO I ANALYZER 400-V
DATE: 1-3-64
COUNTING TIME: 100 MIN
COUNT RATE-CPM
0
100
200
300
400
500
600
CHANNEL NUMBER
0
1
2
3
4
5
6
7
8
9
10
11
12
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15
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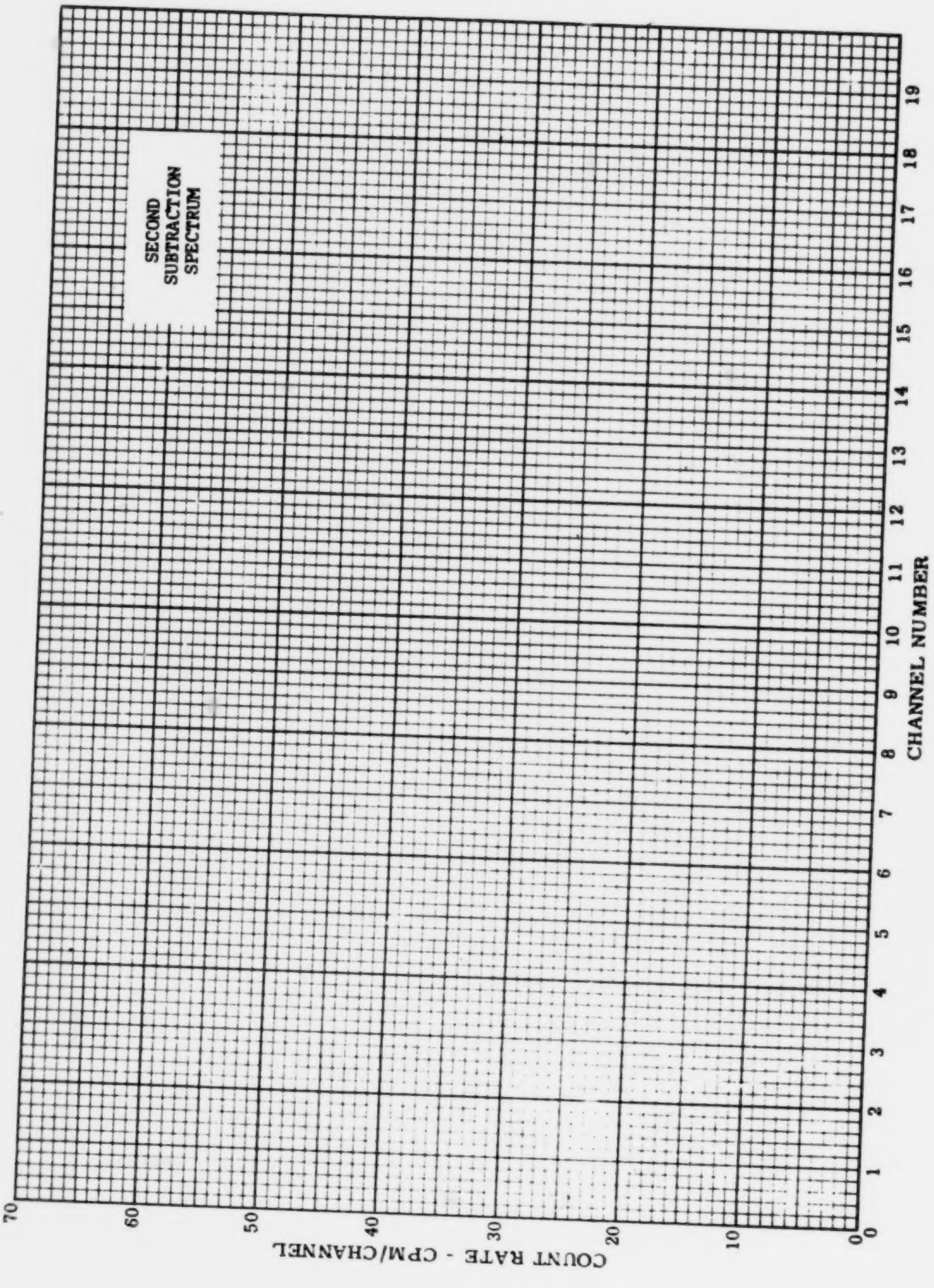
18
19
20

FIGURE 4


SECOND SUBTRACTION SPECTRUM
CHANNEL NUMBER
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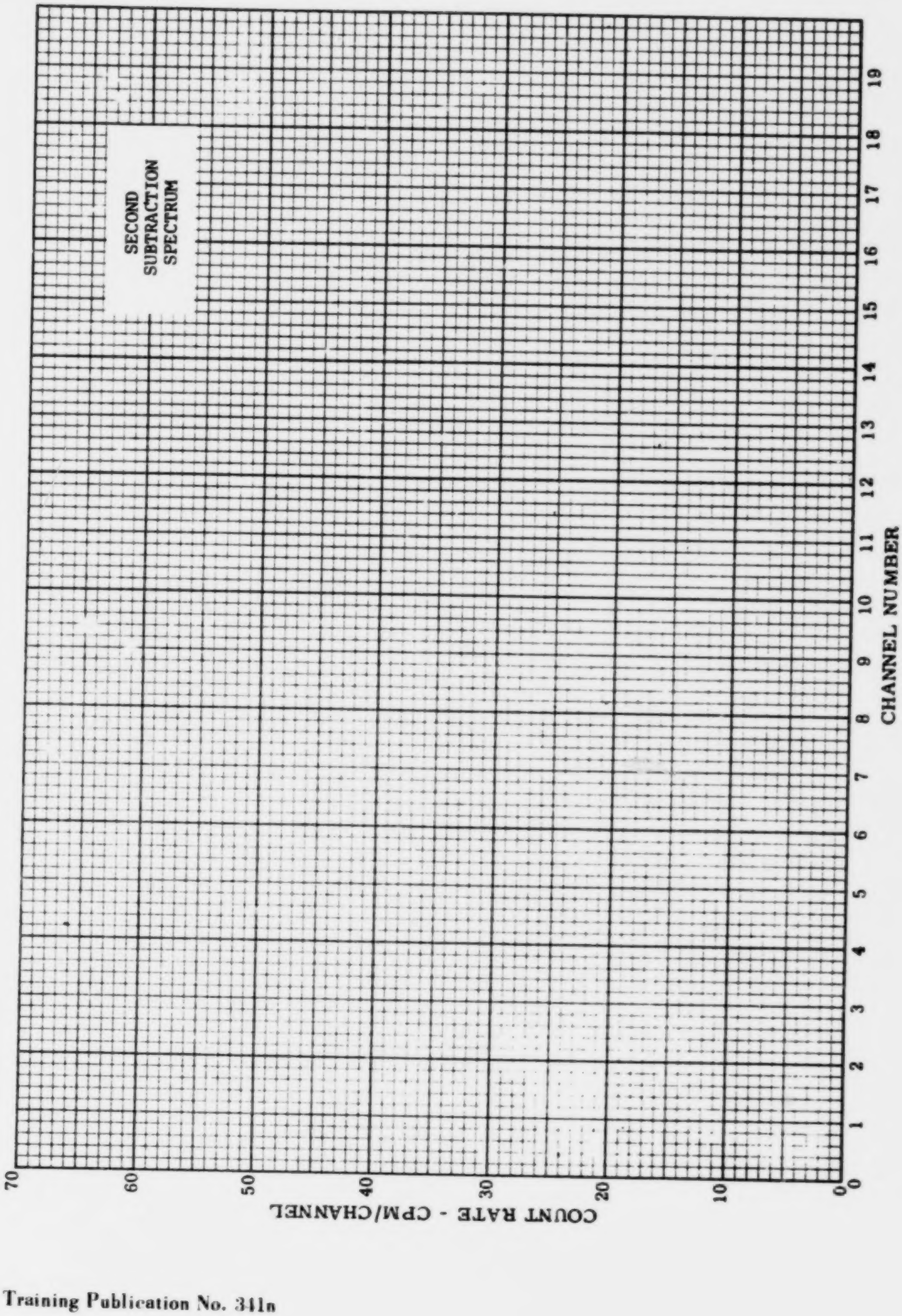
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
COUNT RATE - CPM/CHANNEL
0
10
20
30
40
50
60
70


SECOND
SUBTRACTION
SPECTRUM
COUNT RATE - CPM/CHANNEL
0
10
20
30
40
50
60
70
CHANNEL NUMBER
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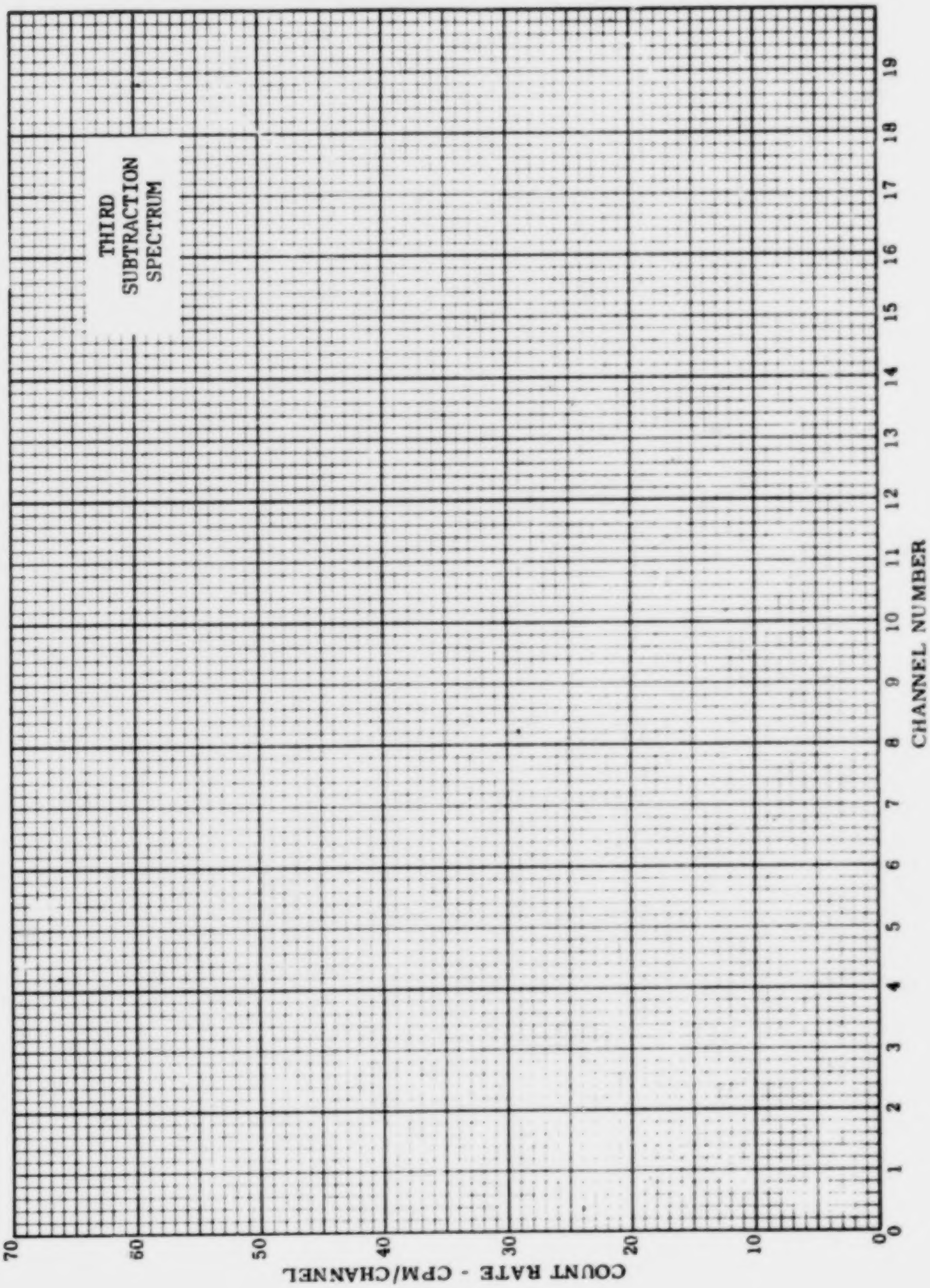
17
18
19


THIRD
SUBTRACTION
SPECTRUM
CHANNEL NUMBER
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2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
COUNT RATE - CPM/CHANNEL
0
10
20
30
40
50
60
70

Analysis of Airborne Particulates

(Laboratory)

PURPOSE

To provide a practical exercise in the qualitative and quantitative analysis of a gamma spectrum obtained by counting a glass fiber filter mat following a 24-hour sampling period.

II. STATEMENT OF CONDITIONS

An air sample was collected in Snugglesville, U.S.A. during a period of atmospheric weapons testing. There are no other significant sources of airborne radioactivity (reactors, etc.) in the area. The sample was collected on a 4" diameter glass fiber filter mat using a High Volume sampler operating at an average flow rate of 50 cfm. The sample was collected over a 24 hour period from 0900 on 11-27-63 to 0900 on 11-28-63. After allowing approximately 7 days for natural airborne radioactivity to decay, the filter paper was counted on a gamma spectrometer on four dates separated by various time intervals, thus yielding the attached data and spectra.

III. PROBLEM AND ANALYSIS

A. Problem

Identify and determine the activity concentration (pc/M^3) of each gamma emitting nuclide present.

B. Analysis

Use the following stepwise procedure:

1. Determine the apparent age of the activity on 12-5-63 by using the decay method relationship as follows:

$$t_1 = \frac{\Delta t}{\left[\left(\frac{A_1}{A_2}\right)^{0.835} - 1\right]}$$

where

t_1 = apparent age at time of count A_1

A_1 = summation of cpm after decay of natural activity (5-7 days after collection).

A_2 = summation of cpm observed at t days after count A_1

Δt = time period (days) between count A_1 and count A_2.

a. Various combinations of the following data may be utilized for this calculation:

Date	Summation (Channels 6-99)
12-5 -63	31615 cpm
12-12-63	24924 cpm

Date	Summation (Channels 6-99)
12-19-63	20311 cpm
1-2 -64	14279 cpm

NOTE: A useful relationship to calculate apparent age on 12-5-63 which may be needed is $At^{1.2}$ = constant.

b. Apparent Age on 12-5-63 = ______days.

2. Using the Fission Product Abundance vs. Time plot, list (in Table 1) all radionuclide possibilities with greater than 0.5% radionuclide abundance.

3. Using the "Gamma Spectrum Catalogue" and/or the Seaborg Tables, list (in Table 1) the predominant gamma energies and half-lives of the radionuclide possibilities.

Table 1. Data obtained from apparent age

Radionuclide possibilities	Predominant gamma energies of possibilities	Half-life of possibilities	Remarks

4. Using the stated channel coefficient (20 Kev per channel), list (in Table 2) energies of major photopeaks observed in each of the four sample spectra.

Table 2. Data obtained from sample spectra

Major photopeak energies observed	Determined half-life of photopeak

5. Using the Compton Continuum subtraction method, and by either summing selected channels in photopeaks (as indicated in Table 3) or by using the peak channel, determine the apparent half-life of the following photopeaks and list in Table 2. (Use Table 3 for calculations and plot on 2-cycle semi-log graph paper.

6. Make a comparison of results obtained in Table 1 and Table 2 by using Table 4; identify nuclides present where possible.

7. Beginning with the 1.6 Mev peak, strip the identified nuclide with this peak from the 12-5-63 spectrum. Using the counting data in the Gamma Spectrum Catalogue for the identified nuclide, perform the following steps:

a. Determine spectrum multiplier

b. Using Table 5, multiply channel-by-channel values from standard spectrum by S.M., and then subtract these adjusted spectrum values from the composite spectrum values, thus obtaining "First Subtraction Spectrum".

c. Plot First Subtraction Spectrum on 4-cycle semi-log paper. Note any uncovered peaks.

8. Determine the activity concentration $\frac{pc}{M^3}$ of the spectrum stripped and record in Table 6.

9. Perform systematic spectrum stripping of other identified nuclides by repeating steps 7, 8, and 9, above for the appropriate energy, channel numbers, and subtraction spectrum. Note and take into account any uncovered peaks.

Table 3. Apparent half-life data

Photo-peak energy	Ch. No.	12-5-63			12-12-63			12-19-63			1-2-64		
		Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm
1.6 Mev	77												
	78												
	79												
	80												
	81												
	82												
	83												
			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$	
0.77 Mev	36												
	37												
	38												
	39												
	40												
	41												
			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$	
0.50 Mev	23												
	24												
	25												
	26												
	27												
			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$	
0.16 Mev	7												
	8												
	9												
			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$			Net cpm $\Sigma =$	

c. Plot First Subtraction Spectrum on 4-cycle semi-log paper. Note any uncovered peaks.

8. Determine the activity concentration $\frac{pc}{M^3}$ of the spectrum stripped and record in Table 6.

9. Perform systematic spectrum stripping of other identified nuclides by repeating steps 7, 8, and 9, above for the appropriate energy, channel numbers, and subtraction spectrum. Note and take into account any uncovered peaks.

Table 3. Apparent half-life data

Photo-peak energy	Ch. No.	12-5-63			12-12-63			12-19-63			1-2-64		
		Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm	Orig. cpm	C. con. cpm	Net cpm
1.6 Mev	77												
	78												
	79												
	80												
	81												
	82												
	83												
			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =	
0.77 Mev	36												
	37												
	38												
	39												
	40												
	41												
			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =	
0.50 Mev	23												
	24												
	25												
	26												
	27												
			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =	
0.16 Mev	7												
	8												
	9												
			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =			Net cpm Σ =	

Table 4. Compilation of information

From spectral data Table 2		From apparent age Table 1		
Observed photopeak energy	Determined half-life	Radionuclide possibilities with that energy	Actual half-life of possibilities	Confirmation

Table 5. Data obtained from spectrum stripping

Channel No.	First Subtraction Spectrum	Second Subtraction Spectrum	Third Subtraction Spectrum	Fourth Subtraction Spectrum
06				
07				
08				
09				
10				
11				
12				
13				
14				
15				
16				
17				
18				
19				
20				
21				
22				
23				
24				
25				
26				
27				
28				
29				
30				
31				
32				
33				
34				
35				
36				
37				
38				

Table 5. Data obtained from spectrum stripping--Cont.

Channel No.	First Subtraction Spectrum	Second Subtraction Spectrum	Third Subtraction Spectrum	Fourth Subtraction Spectrum
39				
40				
41				
42				
43				
44				
45				
46				
47				
48				
49				
50				

Table 6. Final results

Radionuclide identification	Spectrum multiplier	Activity (pc in sample on 12-5-63)	Activity concentration (pc/M^3) 12-5-63

DATA SHEET

SAMPLE: Simulated Air Sample
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M^3
ANALYZER: 400-V
COUNTING TIME: 100 minutes

December 5, 1963 0900 (+ 7 days)
20 Kev per channel
Summation = 31615 cpm
RHINO I

Channel No.	Net cpm per channel	Channel No.	Net cpm per channel	Channel No.	Net cpm per channel
06	2023	38	769	70	30
07	2257	39	790	71	26
08	2716	40	643	72	25
09	1724	41	427	73	22
10	1320	42	265	74	24
11	1242	43	167	75	28
12	1115	44	115	76	45
13	978	45	94	77	68
14	884	46	89	78	106
15	844	47	81	79	136
16	901	48	69	80	156
17	886	49	54	81	152
18	775	50	49	82	124
19	589	51	49	83	88
20	470	52	43	84	58
21	452	53	46	85	36
22	483	54	50	86	28
23	624	55	49	87	21
24	837	56	54	88	16
25	857	57	51	89	13
26	693	58	49	90	13
27	507	59	49	91	11
28	370	60	49	92	12
29	266	61	49	93	10
30	208	62	52	94	10
31	182	63	52	95	10
32	164	64	51	96	8
33	153	65	50	97	8
34	170	66	49	98	8
35	239	67	45	99	8
36	379	68	40		
37	586	69	34		

DATA SHEET

SAMPLE: Simulated Air Sample
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M^3
ANALYZER: 400-V
COUNTING TIME: 100 minutes

December 12, 1963 0900 (+ 14 days)
20 Kev per channel
Summation = 24924 cpm
RHINO I

Channel No.	Net cpm per channel	Channel No.	Net cpm per channel	Channel No.	Net cpm per channel
06	1603	38	703	70	21
07	1802	39	715	71	18
08	2186	40	568	72	17
09	1342	41	360	73	15
10	1031	42	212	74	16
11	986	43	125	75	19
12	889	44	100	76	31
13	671	45	66	77	47
14	692	46	61	78	72
15	652	47	55	79	93
16	681	48	47	80	107
17	650	49	37	81	104
18	558	50	34	82	85
19	438	51	34	83	60
20	369	52	29	84	40
21	360	53	31	85	25
22	382	54	34	86	19
23	489	55	34	87	14
24	654	56	37	88	11
25	671	57	35	89	9
26	395	58	34	90	9
27	395	59	34	91	8
28	290	60	34	92	8
29	211	61	34	93	7
30	166	62	36	94	7
31	145	63	36	95	7
32	129	64	35	96	5
33	122	65	34	97	5
34	141	66	34	98	5
35	206	67	31	99	5
36	339	68	27		
37	533	69	23		

DATA SHEET

SAMPLE: Simulated Air Sample
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M^3
ANALYZER: 400-V
COUNTING TIME: 100 minutes

December 19, 1963 0900 (+ 21 days)
20 Kev per channel
Summation = 20311 cpm
RHINO I

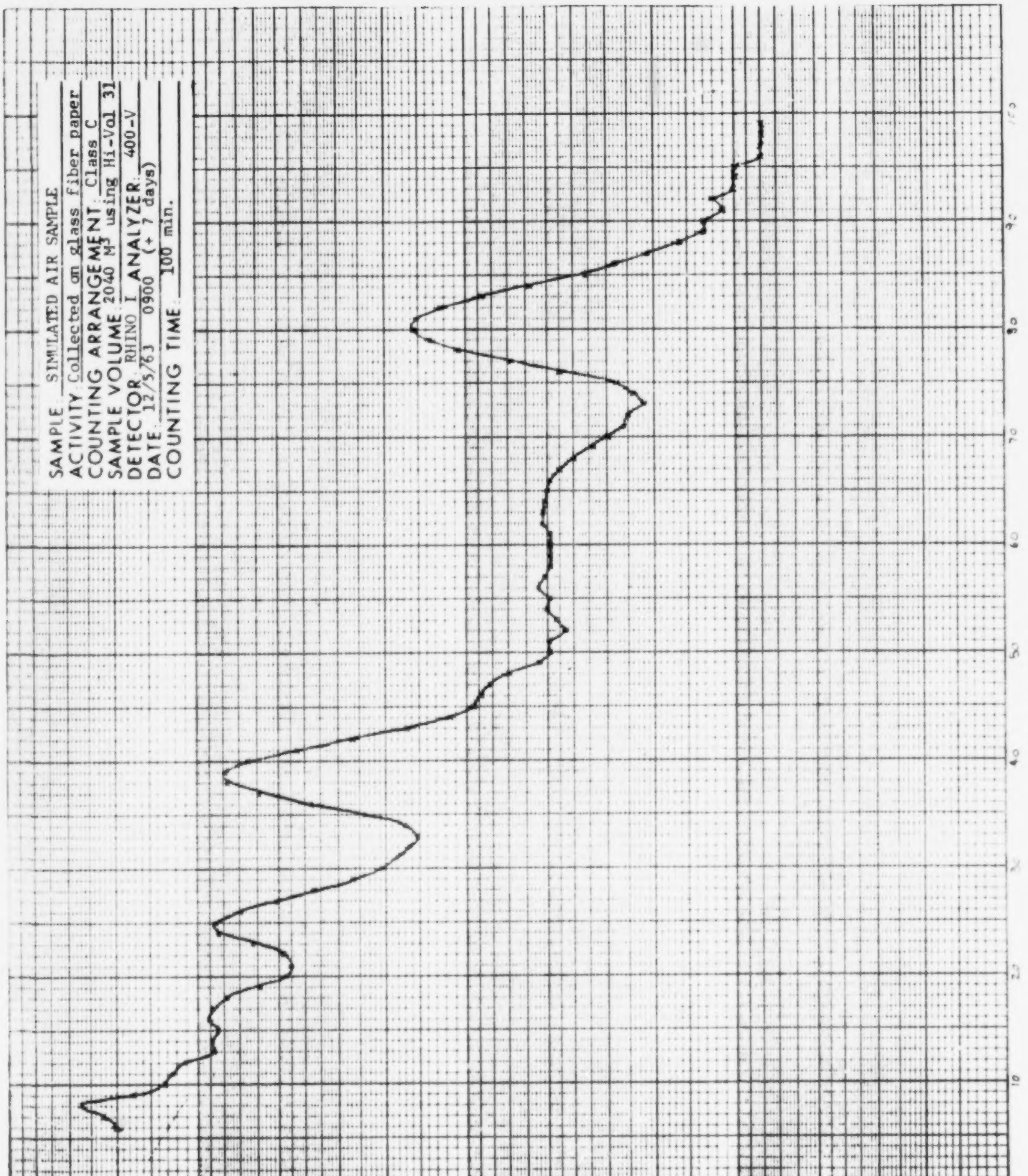
Channel No.	Net cpm per channel
06	1302
07	1471
08	1801
09	1075
10	833
11	811
12	832
13	547
14	560
15	519
16	530
17	495
18	421
19	341
20	299
21	294
22	309
23	393
24	523
25	540
26	436
27	314
28	233
29	171
30	136
31	119
32	107
33	102
34	120
35	182
36	310
37	493
38	649
39	655
40	509
41	310
42	173
43	95
44	60
45	45
46	42
47	38
48	32
49	25
50	23
51	23
52	20
53	22
54	23
55	23
56	25
57	24
58	23
59	23
60	23
61	23
62	24
63	24
64	24
65	23
66	23
67	21
68	19
69	16
70	14
71	12
72	12
72	10
73	11
75	13
76	21
77	32
78	50
79	64
80	73
81	71
82	58
83	41
84	27
85	17
86	13
87	10
88	7
89	6
90	6
91	5
92	6
93	5
94	5
95	5
96	4
97	4
98	4
99	4

DATA SHEET

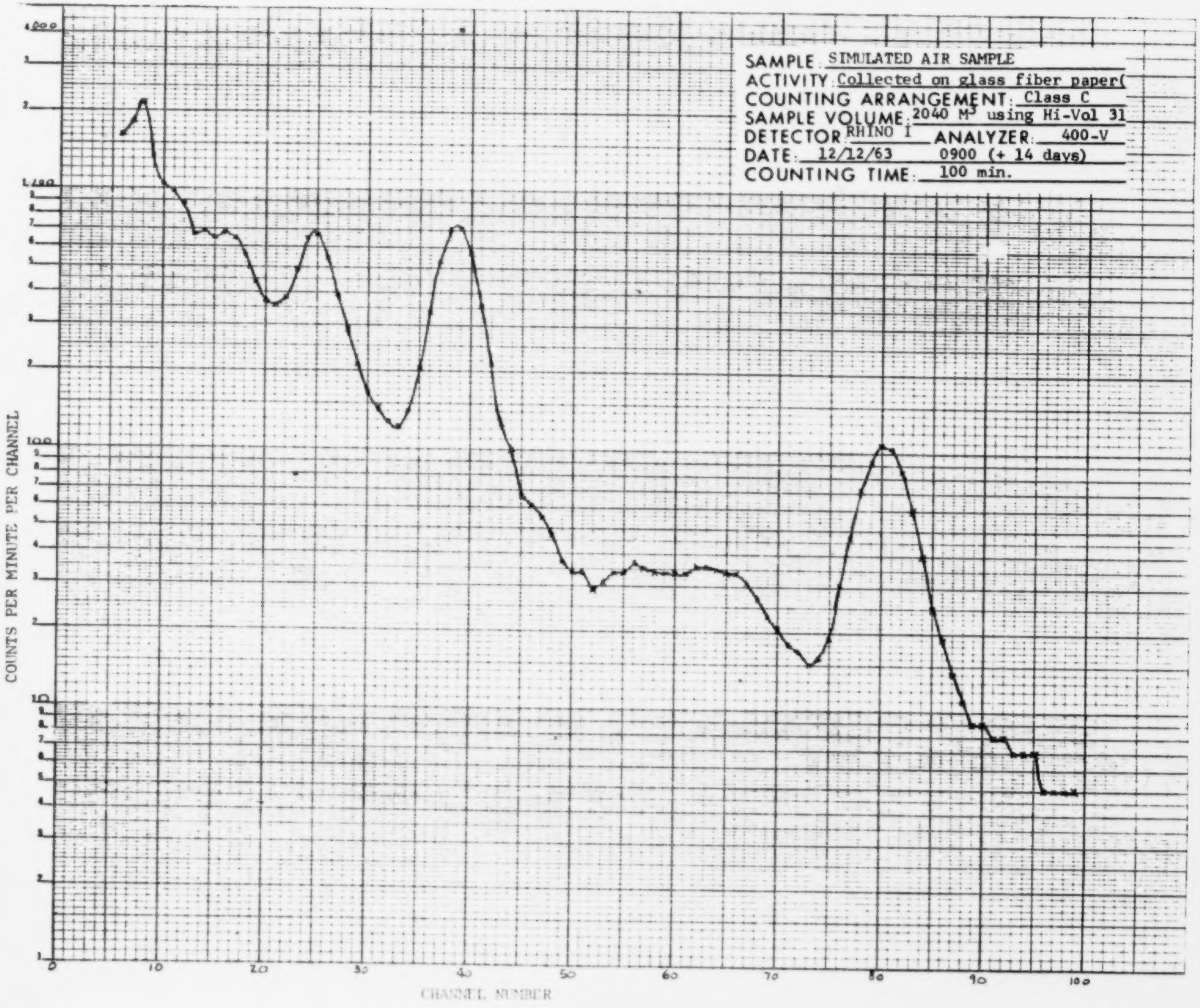
SAMPLE: Simulated Air Sample
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M^3
ANALYZER: 400-V
COUNTING TIME: 100 minutes

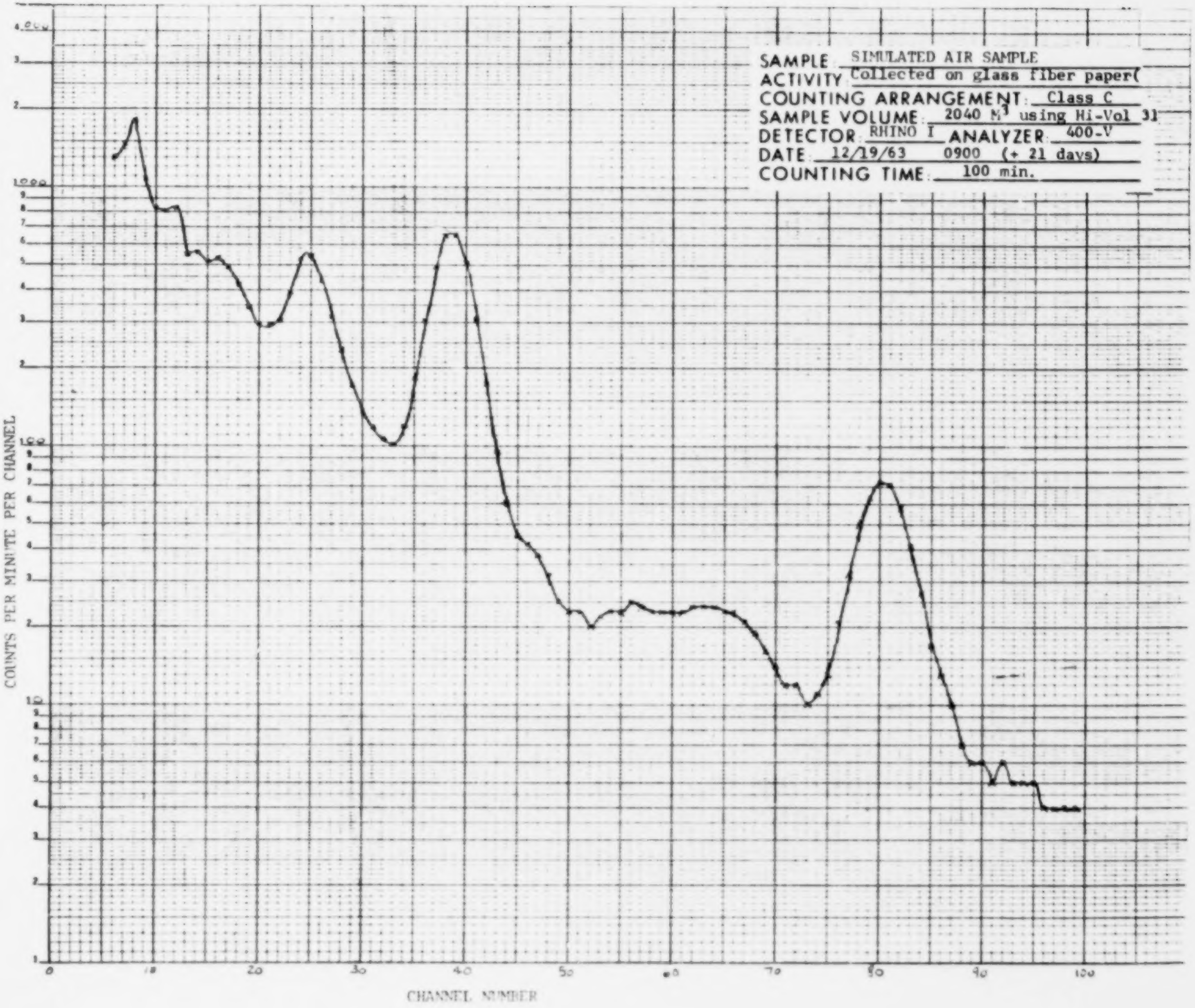
January 2, 1964 0900 (+ 35 days)
20 Kev per channel
Summation = 14279 cpm
RHINO I

Channel No.	Net cpm per channel	Channel No.	Net cpm per channel	Channel No.	Net cpm per channel
06	909	38	562	70	7
07	1043	39	558	71	6
08	1276	40	420	72	6
09	743	41	242	73	5
10	589	42	124	74	5
11	588	43	61	75	6
12	533	44	33	76	10
13	381	45	23	77	15
14	398	46	20	78	23
15	360	47	18	79	30
16	351	48	15	80	34
17	316	49	12	81	33
18	272	50	11	82	27
19	233	51	11	83	19
20	215	52	9	84	13
21	213	53	10	85	8
22	222	54	11	86	6
23	273	55	11	87	5
24	354	56	12	88	4
25	368	57	11	89	3
26	299	58	11	90	3
27	215	59	11	91	2
28	163	60	11	92	3
29	122	61	11	93	2
30	97	62	11	94	2
31	85	63	11	95	2
32	76	64	11	96	2
33	74	65	11	97	2
34	93	66	11	98	2
35	147	67	10	99	2
36	260	68	9		
37	425	69	7		

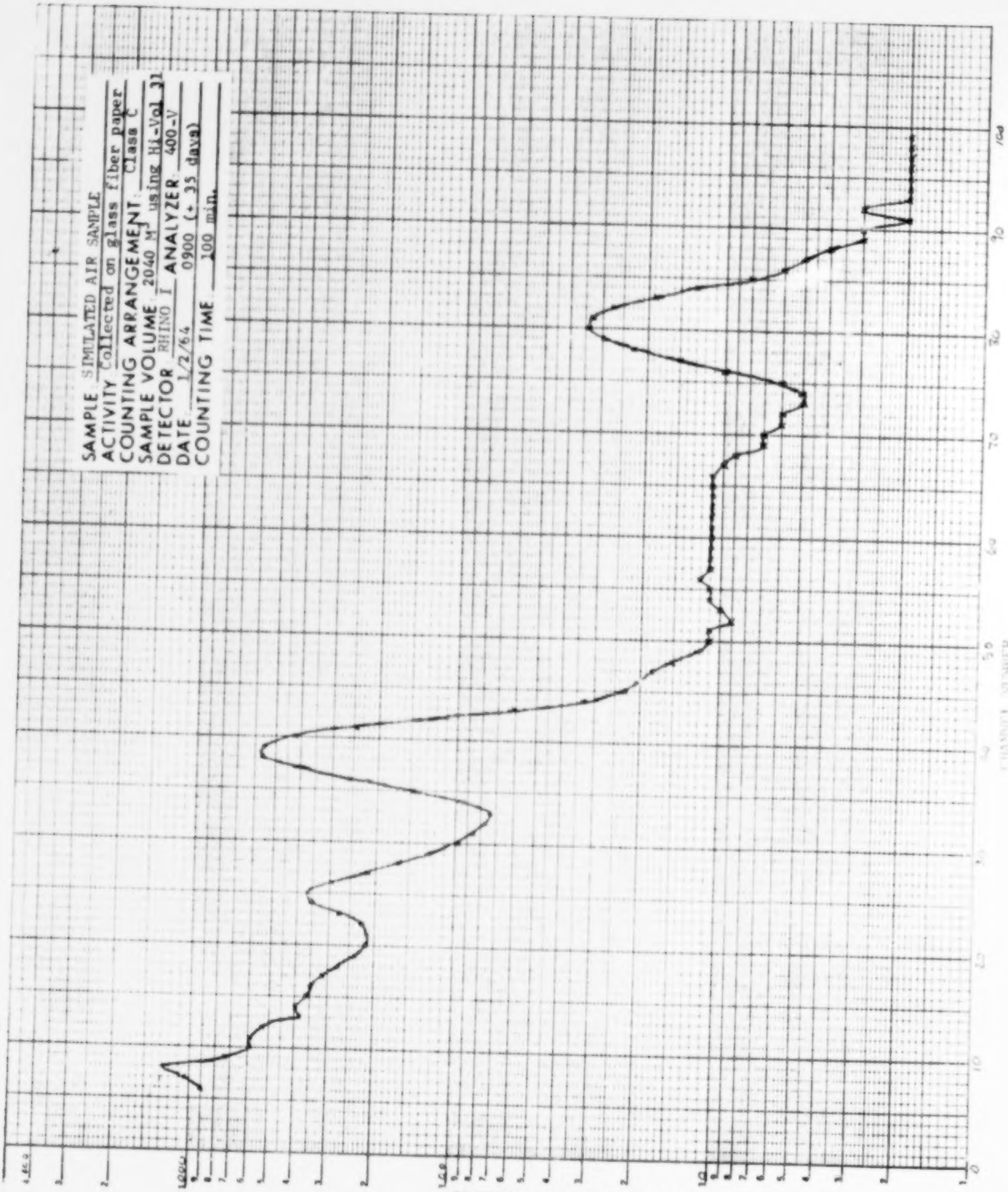


SAMPLE SIMULATED AIR SAMPLE
ACTIVITY Collected on glass fiber paper
COUNTING ARRANGEMENT Class C
SAMPLE VOLUME 2040 M^3 using Hi-Vol 31
DETECTOR RHINO I ANALYZER 400-V
DATE 12/5/63 0900 (+ 7 days)
COUNTING TIME 100 min.


SAMPLE: SIMULATED AIR SAMPLE
ACTIVITY: Collected on glass fiber paper(
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M³ using Hi-Vol 31
DETECTOR: RHINO I ANALYZER: 400-V
DATE: 12/12/63 0900 (+ 14 days)
COUNTING TIME: 100 min.

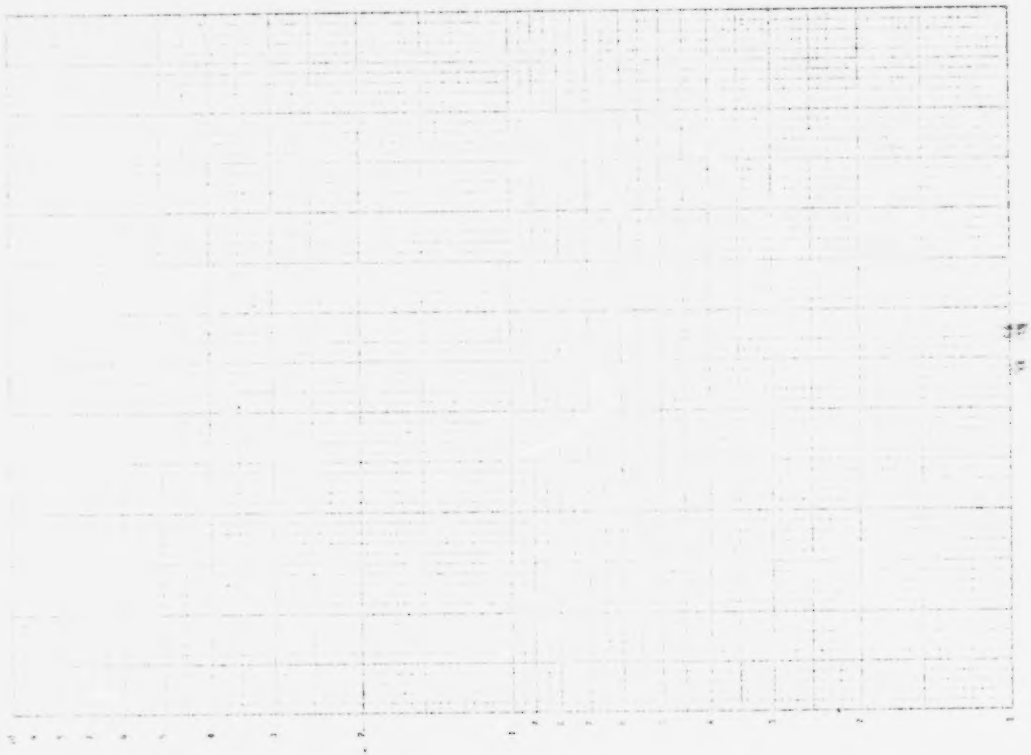


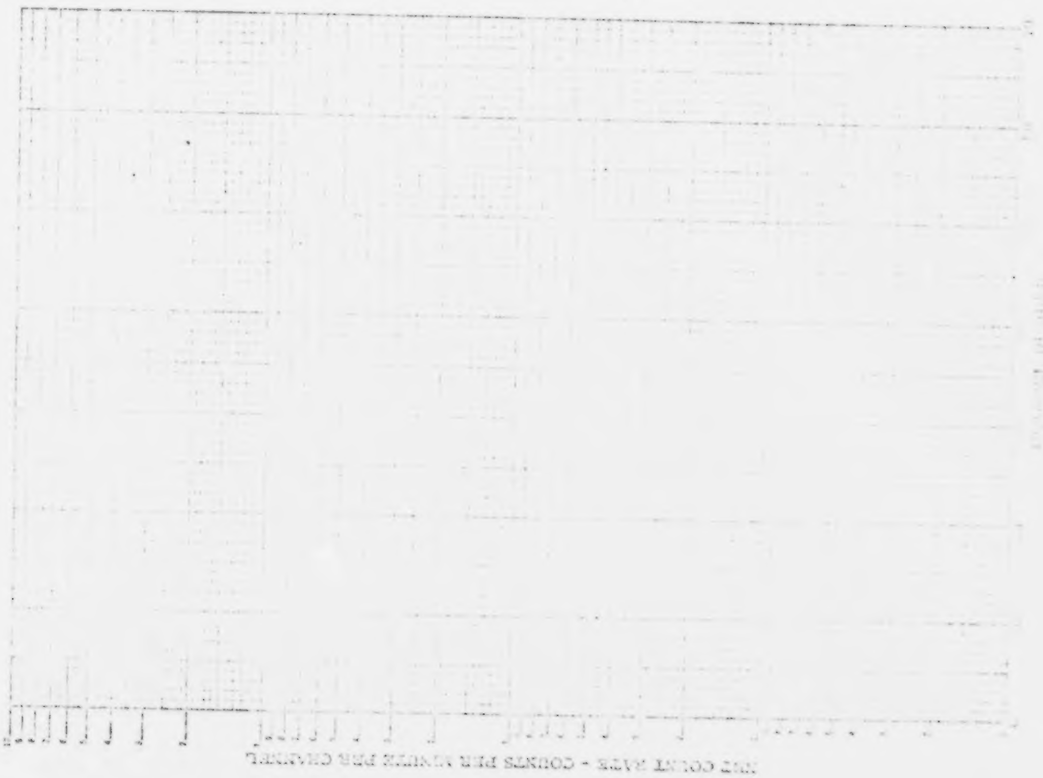
SAMPLE SIMULATED AIR SAMPLE
ACTIVITY Collected on glass fiber paper(
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M³ using Hi-Vol 31
DETECTOR: RHINO I ANALYZER: 400-V
DATE: 12/19/63 0900 (+ 21 days)
COUNTING TIME: 100 min.

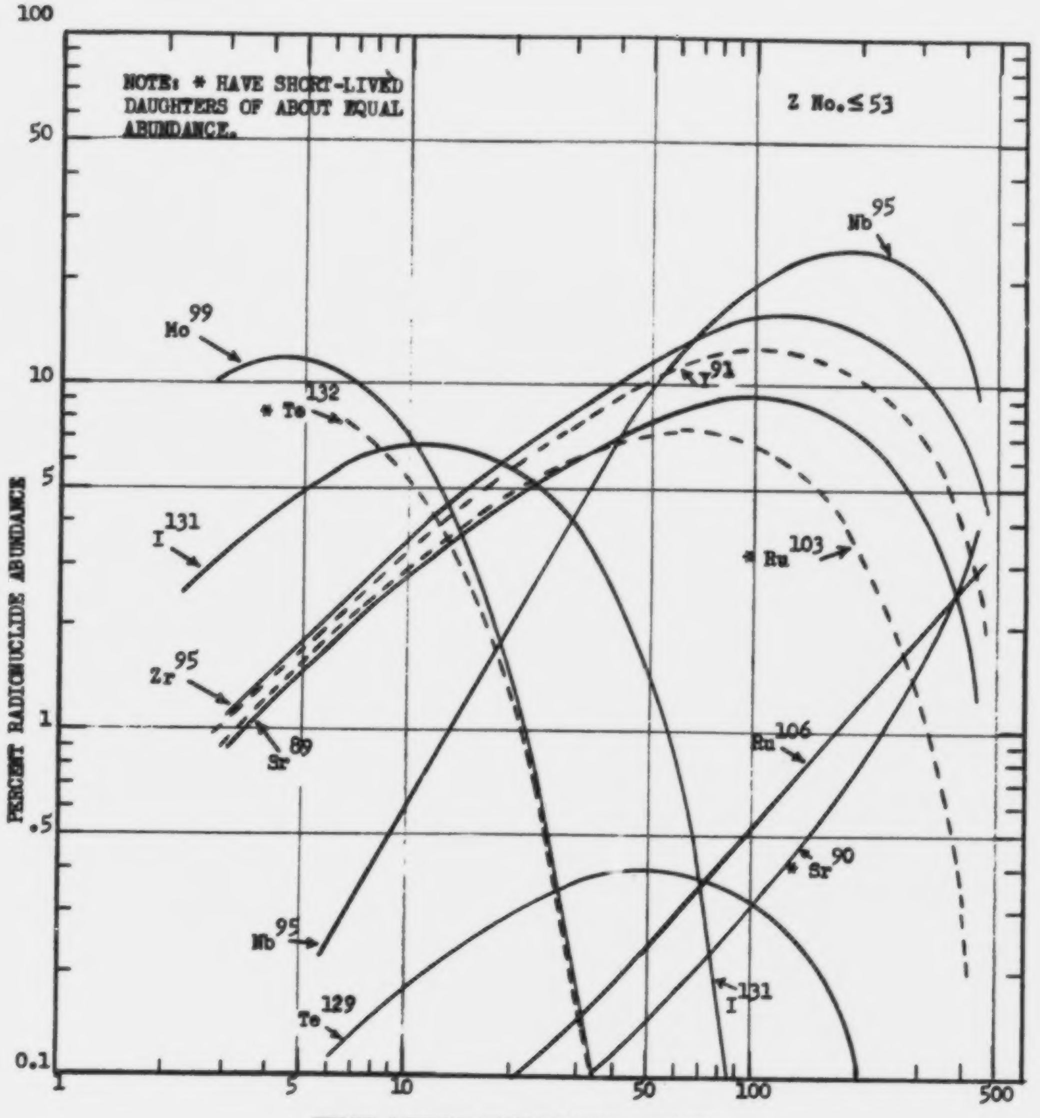


SAMPLE: SIMULATED AIR SAMPLE
ACTIVITY: Collected on glass fiber paper
COUNTING ARRANGEMENT: Class C
SAMPLE VOLUME: 2040 M³ using Hi-Vol 31
DETECTOR: RHINO I ANALYZER: 400-V
DATE: 1/2/64 0900 (+ 35 days)
COUNTING TIME: 100 min.

Division of Radiological Health
3529

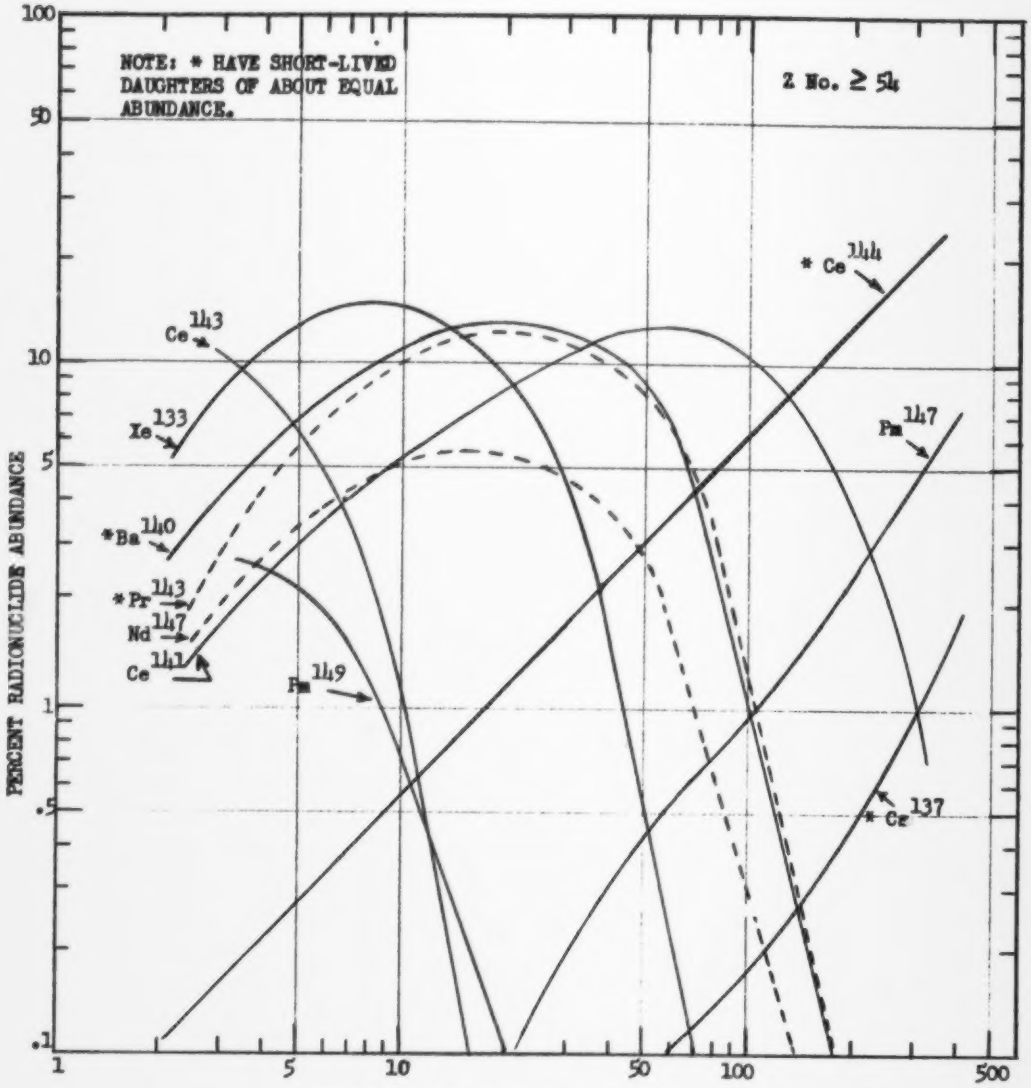



NET COUNT RATE - COUNTS PER MINUTE PER CHANNEL



Relative Abundances of Fission Product Nuclides Up to 500 Days After Fissioning

Figure 2



TIME AFTER FISSIONING - DAYS

Relative Abundances of Fission Product Nuclides
Up to 500 Days After Fissioning

Figure 2a

GPO 886-425

Development of Milk Matrix
(Laboratory)

I. OBJECT

To acquaint trainees with a procedure for quantitative analysis of gamma spectra through the utilization of simultaneous equations.

II. INTRODUCTION

An important fundamental property of gamma spectra is that any portion of the spectrum is proportional to the quantity of activity in the sample. Also the shape of the spectrum is independent of the amount of activity. Therefore, any portion of the curve may be fractionally related to any other portion.

These properties make it possible to determine the amount of interference one radionuclide will contribute to the photopeak of another. Simultaneous equations may be set up to relate the interference from each nuclide in a sample to the observed photopeak of any individual nuclide. The solution of these equations yields, for each nuclide, a relationship between the observed count rate in a photopeak and the count rate which is actually contributed by the nuclide which the photopeak represents.

III. PROCEDURE

A. General

A system of four simultaneous equations will be set up using the calibration data from the "Gamma Spectrum Catalog-RHino I". The equations will be solved for the count rates contributed to each photopeak by the nuclide which that photopeak represents. The contributions of I^{131}, Ba^{140}-La^{140}, Cs^{137}, and K^{40} will be considered.

B. Calculation of Interference Factors

From the standard spectra for the above noted radionuclides calculate the interference factors (f_{xy}) of each nuclide to the other nuclides by the general equation:

$$f_{xy} = \frac{\text{cpm in peak channels of nuclide Y contributed by nuclide X}}{\text{cpm in peak channels of nuclide X contributed by nuclide X}}$$

C. Solution of Matrix

1. Set up four simultaneous equations of the form:

$$N_i = I_i + f_{bi}B_b + f_{ci}C_c + f_{ki}K_k$$
$$N_b = f_{ib}I_i + B_b + f_{cb}C_c + f_{kb}K_k$$
$$N_c = f_{ic}I_i + f_{bc}B_b + C_c + f_{kc}K_k$$
$$N_k = f_{ik}I_i + f_{bk}B_b + f_{ck}C_c + K_k$$

where:

subscripts i, b, c, k, denote I^{131}, Ba^{140} - La^{140}, Cs^{137}, and K^{40} respectively.

N_x = Net count rate observed in photopeak channels for nuclide X.

I_i = Net count rate in I^{131} photopeak channels contributed by I^{131}.

B_b = Net count rate in Ba^{140} - La^{140} photopeak channels contributed by Ba^{140} - La^{140}.

C_c = Net count rate in Cs^{137} photopeak channels contributed by Cs^{137}.

K_k = Net count rate in K^{40} photopeak channels contributed by K^{40}.

f_{xy} = Interference factor which expresses the fractional contribution to photopeak channels of nuclide Y by nuclide X.

2. Solve the simultaneous equations set up in part C-1 for I_i, B_b, C_c, and K_k by the method of determinants.

DATA SHEET

SIMULTANEOUS EQUATIONS

CONTRIBUTIONS TO PHOTOPEAK CHANNELS

	CONTRIBUTIONS TO PHOTOPEAK CHANNELS (Net cpm)			
Photopeak Channels	I^{131}	Ba^{140} - La^{140}	Cs^{137}	K^{40}
16-20 (I^{131})				
22-28 (Ba^{140} - La^{140})				
30-36 (Cs^{137})				
70-76 (K^{40})				

DATA SHEET

SIMULTANEOUS EQUATIONS

Interference Factors:

A. I^{131} (obtained from I^{131} standard)

f_{ii} = ____________

f_{ib} = ____________

f_{ic} = ____________

f_{ik} = ____________

B. Ba^{140} - La^{140} (obtained from Ba^{140} - La^{140} standard)

f_{bi} = ____________

f_{bb} = ____________

f_{bc} = ____________

f_{bk} = ____________

C. Cs^{137} (obtained from Cs^{137} standard)

f_{ci} = ____________

f_{cb} = ____________

f_{cc} = ____________

f_{ck} = ____________

D. K^{40} (obtained from K^{40} standard)

f_{ki} = ____________

f_{kb} = ____________

f_{kc} = ____________

f_{kk} = ____________

SYSTEM OF SIMULTANEOUS EQUATIONS

$$N_i = I_i + \underline{\qquad} B_b + \underline{\qquad} C_c + \underline{\qquad} K_k$$

$$N_b = \underline{\qquad} I_i + B_b + \underline{\qquad} C_c + \underline{\qquad} K_k$$

$$N_c = \underline{\qquad} I_i + \underline{\qquad} B_b + C_c + \underline{\qquad} K_k$$

$$N_k = \underline{\qquad} I_i + \underline{\qquad} B_b + \underline{\qquad} C_c + K_k$$

SOLUTION TO SIMULTANEOUS EQUATIONS

$$I_i = \underline{\qquad} N_i \ \underline{\qquad} N_b \ \underline{\qquad} N_c \ \underline{\qquad} N_k$$

$$B_b = \underline{\qquad} N_i \ \underline{\qquad} N_b \ \underline{\qquad} N_c \ \underline{\qquad} N_k$$

$$C_c = \underline{\qquad} N_i \ \underline{\qquad} N_b \ \underline{\qquad} N_c \ \underline{\qquad} N_k$$

$$K_k = \underline{\qquad} N_i \ \underline{\qquad} N_b \ \underline{\qquad} N_c \ \underline{\qquad} N_k$$

CALCULATIONS

GPO 881-266

Least Squares Quantitative Analysis

Problem Session

I. INTRODUCTION

The purpose of this problem session is to introduce the trainee to the method of least squares analysis of gamma-ray spectra.

II. PROCEDURE

A sample has been run on RHINO I and the data for the first three channels are listed in Table 1. Also listed in Table 1 are the standards for these three channels.

A. Using the basic equation

$$\beta = (A^T wA)^{-1} A^T wp$$

solve for the unknown activities B. Assume that only positive or zero values for B will be accepted.

B. Using the error equation

$$\sigma^2(\beta_i) = C^{-1}(\beta_{ii})$$

where

$$C^{-1} = (A^T wA)^{-1}$$

calculate the 95% confidence intervals for each of the positive or zero values of β.

Table 1

Channel Number	Standard Library (cpm/dpm)			Observed Spectra p (cpm)
	Nuclide X	Nuclide Y	Nuclide Z	
1	1	1	1	2
2	1	2	2	3
3	2	3	0	2